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Glass-Coated Copper as an Architectural Material

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● GLASS AND COPPER have been used in combination forms for ornamental purposes for many centuries. Early artisans found that vitreous enamels could be applied to copper to give pleasing color effects, and such enameled metal has been incorporated into jewelry and objects of art. The cloisonné work of the Chinese and Japanese is based on the enameling of copper and its alloys, and medieval Europeans had similar arts employing combinations of copper and transparent, translucent, or opaque glasses.

To a minor degree, glass-coated copper has been utilized in architecture, but the emphasis has usually been on ornamentation rather than employment of the coating to solve a practical difficulty, namely the tarnishing and oxidation that copper undergoes on prolonged weathering. Yet some technologists believe that thin glass films over copper, brass, and bronze may prove to be a practical way to protect the finishes of these metals and to make possible architectural materials of great utility, durability, and lasting beauty. The purpose of this article is to explore this possibility and to point up some of the problems that stand in the way of development of glass-coated copper for architectural and building purposes.

The Reason for Glass

Copper and its alloys are ideal for building because of their resistance to atmospheric corrosion. But while they do not corrode away, they tarnish on weathering, and this tarnish, except where planned in the original decorative scheme, is considered objectionable. Since the colors of copper and its alloys are especially pleasing, a need exists for a suitable and adequate method for protecting the natural finishes of these metals. It is believed that a vastly expanded use of copper in building would result if tarnish could be prevented.

Lacquers and similar organic coatings are used today, but these provide only temporary protection. They deteriorate with age and weathering and when used on hardware subject to handling, such as brass doorknobs and locks, soon show signs of wear. For endurable protection against tarnish, it is logical to turn to inorganic vitreous coatings which have the proved ability to withstand weathering and wear.

Of the vitreous materials, clear transparent glass is obviously the first choice, since it permits the color of the metal to show through. For variations in artistic effect, however, colored transparent or translucent coatings may be considered, and the color of copper blend-

FIG. 1.

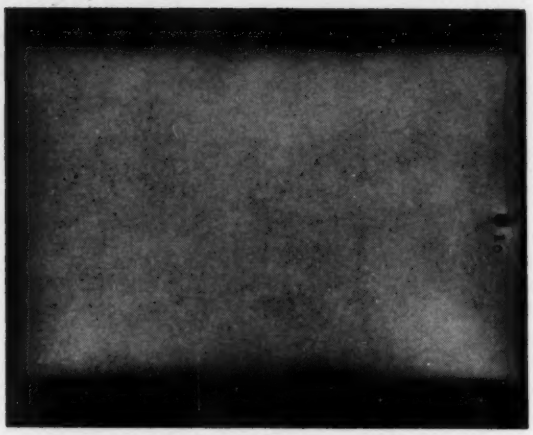

FIG. 1a

These crystal-cote panels from the Battelle study are in perfect condition more than a quarter-century after preparation. Figure 1 has a mottled finish; Figure 1a approximates the natural copper color. Oxide formation at the interface, a factor that promotes adherence, is believed to be responsible for the slight change in color that occurs during firing. Crystal-cote required a firing temperature of 1600°F. for 2½ to 3 minutes. Technologists today believe that equally good, or better, results could be obtained with enamels maturing at 1000°F. to 1200°F.

ing with such coatings is known to give strikingly beautiful surfaces, the like of which can not be produced with any other commercial metal.

Other properties of glass also make it desirable as a coating for copper. Its hardness offers protection against moderate abrasion, it is resistant to atmospheric acids and alkalis, and it is unaffected by water and sun.

FIG. 2. The Parish House of Christ's Church in Rye, New York utilizes vitreous-coated copper for decorative effect. There are 675 enameled copper panels in the spire, steeple, and spandrel trim of this structure. Colors range from deep blue at the base of the buttress to gold at the apex of the steeple.

Problems in Glass-Coating

The main problems inherent in the use of glass as a coating for copper are:

(1) Adherence.
(2) Chipping, cracking, and crazing.
(3) Maintenance of transparency.
(4) The temperatures required to mature the enamel.

Of these, the first three, which to some extent are interrelated, are not too severe when the underlying metal is unalloyed copper; they have largely been solved through past research. The fourth problem area, however, is a source of difficulties which will require much ingenuity to master.

Adherence is related to the surface condition of the metal, the chemical bonds at the interface, and the cubical coefficient of expansion and elasticity of the glasses used. Clean metal and metal with an etched surface promote adherence. Effective cleaning can readily be achieved by washing with detergents, vapor degreasing, cathodic electrocleaning, or sandblasting. Pickling gives a satisfactory surface etch, which is not required if the metal is sandblasted. In general, a better bond is obtained with a rough surface than with a smooth metal face. There is disagreement on the nature of this bond, but one factor is the attractive force between atoms of the base metal and metallic ions in the enamel.

Problems of adherence caused by differences in the cubical coefficients of expansion of copper and glass can largely be obviated by choosing glasses that will be under compression stress on cooling. The amount of compression required depends on the thickness of the coating and of the metal. Modern enameling technologists do not consider thermal expansion a serious problem except when an appreciable volume change

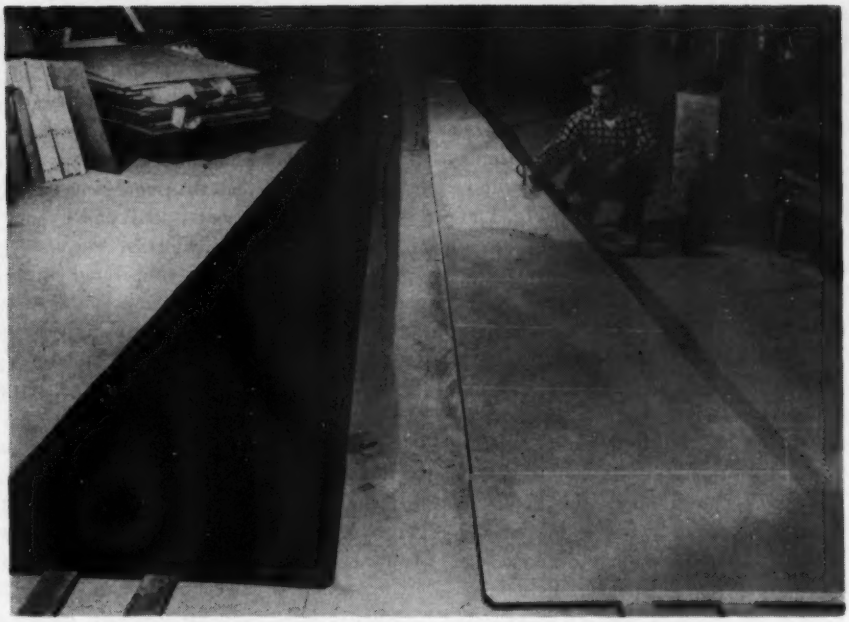

FIG. 3. A workman at the Seaporcel Metals' plant in Long Island City, New York, dusts transparent colored frit onto copper panels for the church building. The colors are being carefully matched to those of panels already fired (on the left). The panels shown here were for the spire and steeple.

takes place in the temperature range where the coating is rigid. Adherence can be improved further by the use of uniform thin coatings, since such coatings are more elastic.

Chipping, cracking, and crazing are to a large extent controlled by the factors that promote adherence. A highly elastic glass in a thin film may withstand severe mechanical deformation. Chipping and cracking are of vital importance if it is necessary to cut, drill, or otherwise fashion the metal after glazing.

Transparency depends on the glass composition, the reactions that occur at the glass-metal interface, the uniformity and thickness of the coating, and whether or not volatile materials are evolved during the firing of the frit. Any oxidation of the metal during firing will cause a deviation from the true metal color, yet, as many past research studies have shown, for best adherence it is desirable that the enamel at the interface be saturated with an oxide of the base metal. Unless the coating is perfectly uniform, smooth, and bubble-free, there will be diffraction of light and a lessening of transparency. Volatilization, particularly volatilization of zinc, may lead to opacity, even when it is not great enough to destroy adherence.

The temperatures required to mature the enamels that have been tried for this purpose are the biggest drawback in the development of glass-coated copper products as architectural materials. For the coated product to be useable as sheet material, it is desirable that the copper be cold-rolled and in as hardened a condition as possible; otherwise the coated material would be easily deformed during handling and in use, even though it be stiffened by crimping or backing. (In addition, cold rolling gives copper a uniform surface finish.) Also, it is desirable that coated extruded shapes be as rigid as possible to provide structural strength.

The maintenance of rigidity, strength, and hardness in glass-coated copper, however, is difficult or impossible under prevalent enameling practices since the glass frit must be heated above the annealing temperature of the metal to achieve coalescence. Even more serious is the temperature factor when attempt is made to glass-coat brass or zinc-containing bronze, inasmuch as diffusion or volatilization of the zinc may occur with the destruction of adherence and the transparency of the coating.

Methods for obviating the difficulties caused by enamel-maturing temperatures have been suggested, and these will be discussed later.

The "Crystal-Cote" Process

Possibly the most comprehensive research done on the vitreous coating of copper was conducted at Battelle Memorial Institute by Sullivan, Simpson, Witschey, Gillett, Bole, and Harder in the early thirties. This work, sponsored by the Copper and Brass Research Association, was summarized in a report made to that Association, dated November 30, 1934, and was also the basis for a series of patents, the first of which, U.S. Patent 1,934,788, entitled "Sheet Metal Faced Structural and Insulating Unit," was issued to G. A. Bole and J. D.

Sullivan on November 14, 1933. The study covered the enameling of copper both with clear and opaque compositions, and the clear glazes that were developed and proved successful in laboratory and exposure tests were called "crystal-cote" enamels. Test panels coated with these enamels prior to 1934 are still in a perfect state of preservation, and after more than a quarter century retain the original color without blemish or without defect in the coating. The panels illustrated in Fig. 1 are from a collection that was maintained in an interior environment, the outdoor exposure panels having been discarded.

The Battelle work of that period, although it never led to commercial glass-coated copper products, is of interest, inasmuch as it points up both the successes and the problems that are encountered in any attempt to develop glass-coated copper architectural and building products.

At that time, little was known about the nature of adherence and the metal-to-metal bonds at the enamel-metal interface, and it was thought that difference in thermal expansion was the critical problem. Battelle researchers, to get around the problem, selected glass compositions having what they called high "stretch factors," or "ability to withstand stretch without rupture." Some thirty frits were considered, seven of which were compounded, smelted, milled, and applied to copper. The most satisfactory was a borosilicate glass, with a composition as shown in Table I. This was the basis for subsequent crystal-cote compositions.

Cleaning was accomplished with wetting agents; cathodic electro-cleaning using a soda ash, trisodium phosphate, and sodium hydroxide electrolyte and iron anodes; vapor degreasing with trichlorethylene and sandblasting—all of which gave satisfactory results. Pickling of the copper panels, other than those sandblasted, was done with dilute nitric acid or with a bichromate-sulfuric solution. The frit, finely ground, was prepared as a slip, with bentonite or ammonium alginate as the floating agent, and sprayed on the metal. The best application proved to be fifteen grams, dry weight, per square foot, one side. Firing of crystal-cote was usually for two-and-one-half to three minutes at 1600°F.

The coated panels prepared in this manner could withstand much distortion without the surface cracking. Impact adherence with thin coatings was good and improved with increases in the thickness of the copper. Even today, more than a quarter century later, one can scrape the glass films from test panels only with great difficulty. Some colorants, added to clear crystal-cote, improved adherence. For instance, a greenish opaque enamel made with additions of chromium oxide gave a coated copper that could be bent at right angles without destroying the bond. Thin strips of this material could even be tied into knots.

In salt-spray tests, running for five hundred and twenty hours, there was no evidence of breakdown of the enamel that would expose the copper surface, although a slight iridescence developed on the surface of the glass. Accelerated outdoor exposure tests after two years gave no evidence of deterioration, crazing, or dulling of the finish on well enameled and fired pieces. The materials also stood up in weatherometer tests involving ultra-violet light, water sprays, and refrigeration; in sulphide atmospheres; under thermal shock tests much more severe than could reasonably be expected in service environments; and under one year of outdoor exposure at Miami, Florida.

Several companies cooperated with Battelle and the Copper and Brass Research Association in the development of crystal-cote copper and in the application of the process to experimental commercial products. Among these were the Ferro Enamel Corporation, Cleveland; the Youngstown Press Steel Company, Warren, Ohio; the Beaver Enameling Company, Ellwood City, Pennsylvania; Vitreous Enameling Company, Cleveland; and Davidson Enamel Products, Inc., Lima, Ohio. A wide variety of products were coated experimentally, including spandrels, window panels, signs, pan and joint strips for ceilings, ridge rolls, shingles, ridge finials, downspouts, light reflectors, switch plates, tiles, ash trays, bowls, mugs, and cooking utensils. Odd shapes, such as screens and coiled tubing, were also coated. The process was used to enamel the strip joints, signs, and other copper work used in the Statue of Liberty tunnels.

Despite the success of the experimental efforts, however, crystal-cote copper did not become an item of commerce. The main reason for this, according to some who followed the development closely, was the fluctuating price of copper in the pre-war period, which did not permit enameling companies to make firm bids. Also there were technical inadequacies. The process could not be extended to the traditional architectural brasses and zinc-containing bronzes. In addition, there was the problem of maintaining rigidity and flatness in copper sheet. At the firing temperature, some annealing occurred with a softening of the metal. Yet in the short time involved in maturing the enamel, the stresses in cold rolled sheet were not relieved uniformly, resulting in warpage. With the shortages of copper that occurred with the outbreak of World War II, there was little incentive for further development.

Obviating the Temperature Problem

Several methods have been suggested for avoiding the softening of copper that occurs during firing of the frit. One is to use precipitation-hardening alloys. In experiments, Battelle found that some alloys of this type, if given a hardening treatment before enameling, were not markedly softened during the firing. Some specimens were fired to mature the enamel and then immediately transferred to a furnace and held at a suitable temperature for precipitation hardening, and after remaining in this furnace for the required time were removed and cooled in air. This arrested cooling frequently increased the hardness of the material substantially, although ad-

Table I*

Raw Batch Composition		Melted	
Ingredient	Weight Grams	Oxide	Weight %
Sodium Nitrate	6.86	Na_2O	2.5
Potassium Nitrate	5.37	K_2O	2.5
Zinc Oxide	2.50	ZnO	2.5
Barium Carbonate	12.90	BaO	10.0
Cryolite	12.50	Cryolite	12.5
Boric Acid	35.40	B_2O_3	20.0
Flint	50.00	SiO_2	50.0
	125.53		100.0

*Composition of the borosilicate glass that gave the best results in the Battelle experiments.

herence was not as good as it was on the straight copper.

Precipitation-hardenable alloys are regarded today as no more than a speculative and probably infeasible solution to the temperature-softening problem. Even if a satisfactory metallurgical product could be made, its cost would be considerably higher than the normal mill product. Also this approach would not solve a major problem — which is how to glass-coat the traditional brasses and commercial bronzes that are favored in architectural and building applications.

A temperature of 1600°F., required for maturing an enamel, is very close to, or even above, the melting point of many of the architectural bronzes and brasses. Thus, yellow brass containing sixty-seven per cent copper and thirty-three per cent zinc, melts at about 1725°F., and Muntz metal (sixty per cent Cu, forty per cent Zn) melts at approximately 1545°F. Obviously these metals cannot be subjected to 1600°F. frit-firing temperatures. Also, all of the commercial brasses, bronzes, and nickel-silvers used as architectural metals contain zinc, ranging from five to more than forty per cent. Zinc volatilizes at 1665°F., and its vapor pressure at 1600°F., is high enough to prevent enamel adherence. Even if adequate adherence could be obtained, as some preliminary work has shown may be possible, the volatilization of zinc from the alloy tends to cause opacity in the glass coating, due to the formation of zinc ozide.

In view of these facts, the possibility of developing glass-coated copper alloys that would have wide architectural acceptance seems to rest primarily on the ability of technologists to come forth with *suitable* low-melting frits or methods of applying molten glass in such a manner that serious softening of the metal and destructive zinc volatilization do not occur. Can this be done? Some authorities say yes; others are doubtful.

There is no question about the availability of low-maturing enamels. Lead-containing enamels mature at much lower temperatures than borosilicate compositions. In the 1930's lead-containing enamels were avoided because of the in-plant health hazard. These can be safely handled today, and present enamels for aluminum contain lead. Other compositions have been suggested. Enamels of lower-maturing temperatures to be *suitable* would also have to give good adherence and would need to be adequately resistive to shock, abrasion, and corrosion to fulfill their protective function. They would also have to remain clear upon application to copper or copper alloys, except in those instances where a desirable opacity or coloring effect were obtained.

Some annealing would be expected even with an enamel maturing at 1000°F., but the strength loss might not be too serious, even for extruded shapes to be used for structural purposes. At the lower firing temperature, less warpage of sheet would be expected, but whether a satisfactorily flat sheet of curtain-wall size could be made remains problematical.

It has been suggested that some method of spraying or rolling molten glass onto copper and its alloys may be a practical approach to the problem, particularly if this can be accomplished without heating the metal appreciably, thus avoiding the annealing effect and zinc volatilization. The possibilities here seem doubtful, in view of the nature of the bond. According to present theory, there must be a reaction between the enamel and the surface to get adherence. This requires heat.

In spite of the unsolved problems, specialists in the enameled metal field are optimistic about the possibility of glass-coated copper architectural materials (Figures 2 and 3). John D. Sullivan, one of the developers of crystal-cote, believes that there is a place in the construction field for enameled copper and that a satisfactory product can be developed using an enamel that matures in the range of 1000°F. to 1200°F. He feels that the annealing effect at lower temperatures should not be so serious as to discount the possibility of an acceptable product and that the warpage problem in sheet material might be solved by coating each side or giving cold-rolled sheet a preliminary anneal to relieve stresses. With lower firing enamels, he believes there may even be a possibility of coating brasses with a zinc content as high as 10 per cent or more. All of these possibilities would, of course, have to be proved through research.

Even at the present state of the art (which could be quickly improved merely by updating a quarter century), there would seem to be plenty of opportunity for developing excellent building materials. For curtain wall and paneling purposes, and possibly for roofing and roof shingles, glass-coated copper sheet — even though softened by annealing — could be backed up with sprayed reinforced plastics or bonded to asbestos, asphalt, or foamed materials to give the needed rigidity. All copper paneling, unless it be in prohibitively costly thickness, would have to have such backing anyway to keep it from "dishing" and to deaden sound. Very thin asphalt-backed, glass-coated copper shingles would possibly give a roof that would last a lifetime in the true or nearly true copper color. The main difficulty would be in handling the softened metal after firing and until it was bonded to the rigidifying backing, but this is merely a mechanical problem. This practical approach for the development of commercial building materials would leave some intriguing technological problems unsolved, but, perhaps while working in this field, a basis of information would be obtained for the eventual mastering of those problems.

Foamed Silica for High Temperature

by *Francis C. Lin* CERAMIC APPLICATION ENGINEER, MATERIALS LABORATORIES, WESTINGHOUSE ELECTRIC CORPORATION, PITTSBURGH 8, PA.

A pure foamed silica was evaluated and found to have a bulk density range from 10 to 60 lbs./cu.ft., a dielectric constant range from 1.3 to 2.1, and a room temperature loss tangent less than 0.0004 at 10,000 mc. These excellent electrical properties, along with high temperature stability, may lead to the use of this material as a substitute for organic dielectrics in high temperature, airborne microwave electronic applications. The physical properties of foamed silica are presented. Methods for making electrical measurements are also discussed.

● RECENTLY, ELECTRONIC COMPONENTS HAVE BEEN REQUIRED to perform complicated functions under higher operating temperatures than those for which they were originally designed. Of primary consideration in the selection of dielectric materials for electronic applications are unique dielectric properties, simplicity of manufacture, and machinability into final size and shape. Plastics such as teflon, polystyrene, polyurethane, and epoxy resins, have been used as insulation in many microwave electronic applications as either solid or foam. The increasing temperature requirements due to higher power output have led to a study of high temperature dielectric materials as a substitute for teflon.

A pure silica foamed dielectric material (such as Pittsburgh-Corning's Foamsil) was suggested as a workable substitute for teflon, since its electrical properties at microwave frequencies are somewhat similar to those of teflon, and since it would operate at much higher temperatures than teflon. A typical requirement is that the material have high temperature (300-500°C) insulation properties such as: a dielectric constant of 2.1, a loss tangent of less than 0.0005, and a volume resistivity of 10^{16} ohm-cm or higher.

During the past ten years, American manufacturers have developed, patented and published processes for making foamed glass from many available commercial glasses. These processes, [1,2,3,4] in general, produced the foamed glass by firing glass powder with a cellulating agent, usually carbon, dolomite ($CaCO_3 \cdot MgCO_3$) or calcium carbonate. The glass fuses and entraps particles of cellulating agents, which at the same time decompose or oxidize. The resulting gas is retained in the glass in many small cells. The preparation of the batch and the heat control at firing temperatures of a given batch composition are extremely critical. As a result, many of these processes were shown to be impractical and thus became obsolete.

An extensive literature search[5,6,7,8,9,10,11,12,13] was made

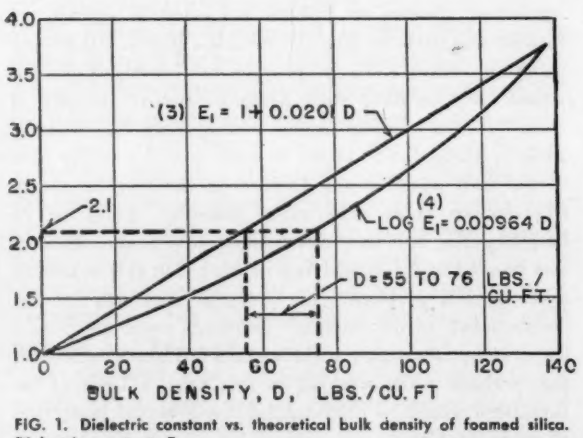

FIG. 1. Dielectric constant vs. theoretical bulk density of foamed silica. Dielectric constant, E_1

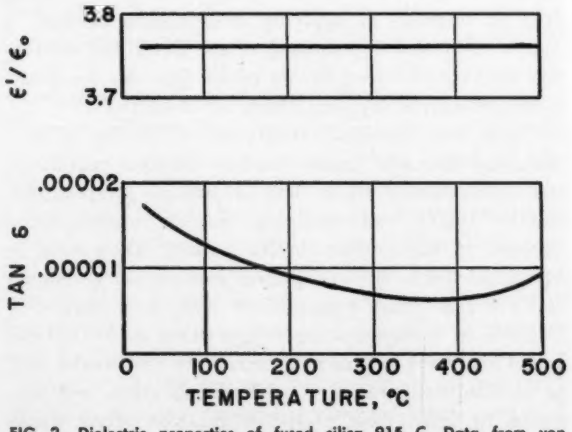

FIG. 2. Dielectric properties of fused silica 915 C. Data from von Hippel[15].

Microwave Electronic Applications

and many European patents were reviewed.[13] It was found that there is no process available to produce a foamed material with silica content higher than 85 per cent by weight, except Pittsburgh-Corning's Foamsil process. Foamsil, although an excellent thermal insulation material, does not have the required electrical properties, because of the size, uniformity, and distribution of its bubbles.

This paper covers the evaluation of the electrical properties of a material similar to Foamsil, but with a controlled density and bubble structure.

Background

A foam may be defined as a combination of solid and gaseous materials of fine and uniform structure. The solid portion is a continuous phase; the gaseous portion may be either discrete or continuous. When the gas is discrete, a closed-cell or unicellular foam will result. When the gas is continuous, the foam is an open-cell type. It is reasonable to assume that the gas has a dielectric constant of 1.00 and that its dielectric loss is negligible. The solid constituent of the foam will have a dielectric constant of say, between 2.0 to 9.0. The dielectric constant and the dissipation factor or the loss tangent of the foam are therefore intermediate between those of the solid and the gas. Two formulas for the dielectric constant of a foam as related to its bulk density have been suggested:[14]

$$E_1 = 1 + K_1 D \quad (1)$$

$$\text{and } \log E_1 = K_2 D \quad (2)$$

where E_1 is the dielectric constant of the foam, K_1 and K_2 are the constants that relate the dielectric constant or the logarithm of the dielectric constant to the bulk density, and D is the bulk density of the foam.

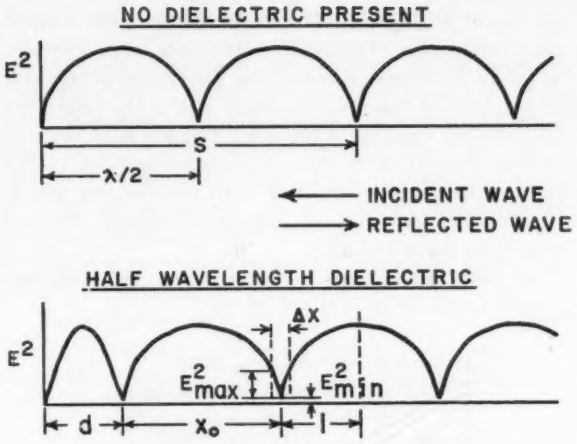

FIG. 5. Standing-wave patterns in wave guide.

For fused silica, let $E_1 = 3.76$ and D = 137.28 lbs/cu ft, the following equations are then obtained:

$$E_1 = 1 + 0.0201\ D \quad (3)$$

$$\log E_1 = 0.00964\ D \quad (4)$$

As illustrated in Figure 1, it is estimated that at E_1 equals 2.1, the bulk density of the foamed silica should lie between 55 and 76 cubic feet.

The loss tangent of fused silica is reported[15] to be less than 0.0002 at frequencies as high as 10,000 megacycles per second and at temperatures between 25 to 500°C as illustrated in Figure 2. It is estimated that the

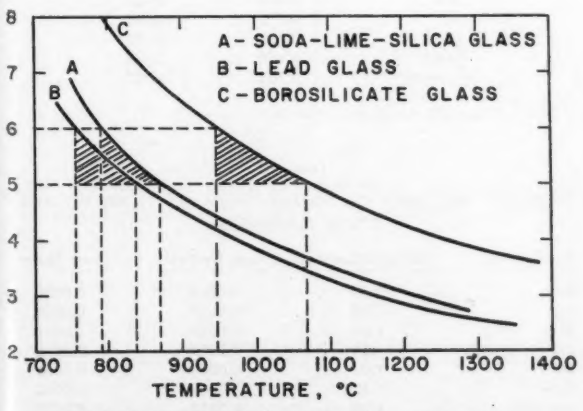

FIG. 3. Viscosity of glasses at various temperatures. Viscosity in poises $\log_{10}$

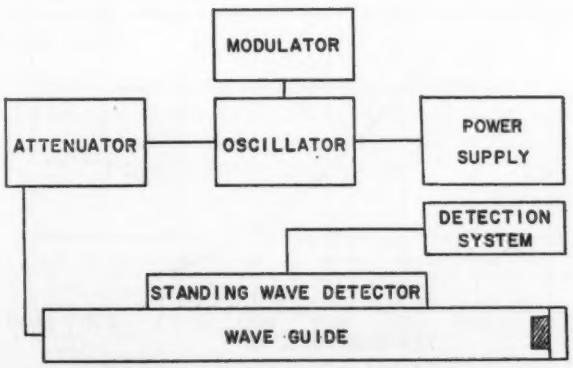

FIG. 4. Electrical system for microwave dielectric measurement.

loss tangent of foamed silica should have a value in the neighborhood of 0.0002. Any value greater than 0.0002 is probably a result of impurities in the silica batch and contamination due to undecomposed cellulating agent.

Experimental Procedures and Results

The first set of experiments were made using glass. Table I shows some typical commercial glass compositions.[16] The viscosity curves of these glasses[17] are shown in Figure 3. Experiments had shown that best foamed glass results were obtained when the glass had a viscosity of 10^5 to 10^6 poises. A cellulating agent such as charcoal or graphite for closed cell structure or $MgCO_3$, $CaCO_3$, or dolomite for open-cell structure is blended with the preground glass. This mixture is then brought up to the foaming temperature and held until the foaming action is completed. A similar process was followed using a pure silica instead of glass.

From these experiments it was obvious that a critical control had to be maintained of the:

1. Percent of cellulating agent
2. Particle size of cellulating agent
3. Particle size of matrix composition
4. Foaming temperature
5. Soaking time
6. Amount of contamination

Table III shows the bulk densities of some of the foamed silica samples.

Microwave Dielectric Measurement

The method of measurement of the dielectric properties of solid dielectrics proposed by Roberts and Von Hippel[15] has been applied using a 3 cm wavelength in a rectangular wave guide. A simplified procedure has been developed by Dakin and Works[18] of the Westinghouse Research Laboratories for calculating the dielectric properties from the measurements of the position of a minimum of the standing wave and the ratio of the minimum field strength to maximum field strength of the

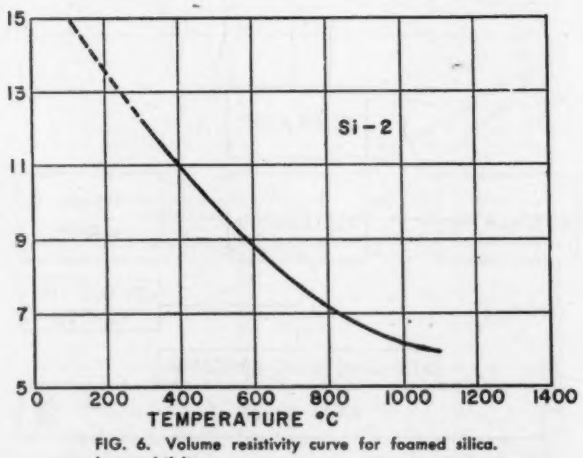

FIG. 6. Volume resistivity curve for foamed silica. Log resistivity

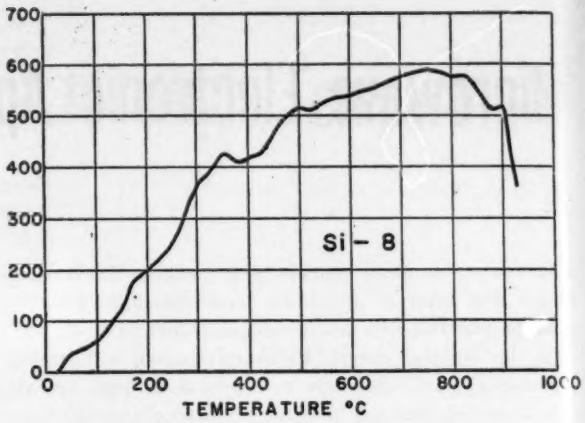

FIG. 7. Thermal expansion of foamed silica. Thermal expansion (10^{-6})

standing wave. This paper was published in the Journa of Applied Physics in 1947.[18] The electrical system o the measuring instrument is shown in Figure 4.

This method consists of reflecting the wave at norma incidence from a slab of the dielectric which is place against a perfectly reflecting surface. The process o reflection sets up standing wave in the space regio in front of the sample as a result of the superpositio

Table I

TYPICAL COMMERCIAL GLASS COMPOSITIONS

Oxide	Soda-Lime-Silica	Lead	Borosilicate
SiO_2	72.7	65.0	81.0
Al_2O_3	1.0	.....	2.0
CaO	6.4	.....	.....
MgO	1.7	.....	.....
PbO	.....	20.0	.....
B_2O_3	1.4	.....	13.0
Na_2O	16.1	7.5	4.0
K_2O	0.7	7.5	.....

Table II

DENSITIES OF FOAMED SILICA SAMPLES

Sample No.	Gm./cc.	Lb./cu. ft.
Si-2	0.774	46.43
Si-5	0.485	30.30
Si-6	0.402	25.05
Si-9	0.376	23.46
Si-11	0.559	34.88
Si-14	0.499	31.14
Foamsil*	0.282	17.37

* Pittsburgh-Corning's experimental material.

Table III

DIELECTRIC PROPERTIES OF FOAMED SILICA AT 10,000 MC AND ROOM TEMPERATURE

Sample No.	Dielectric Constant	Loss Tangent	Loss Factor
Si-2	1.59	0.0005	0.0008
Si-5	1.35	0.0005	0.0007
Si-6	1.40	0.0010	0.0014
Si-9	1.38	0.0007	0.0010
Si-11	1.40	0.0004	0.0006
Si-14	1.45	0.0007	0.0010
Foamsil*	1.32	0.0013	0.0017

* Pittsburgh-Corning's experimental material.

of the incident and reflected waves as illustrated in Figure 5.

The separation of the first minimum from the face of the sample will depend on the wavelength of the wave in the sample and the sample thickness, since the first minimum will be an integral number of halfwaves from th reflecting surface behind the sample. This results in shortening of the wavelength in the sample. Insertion of the dielectric shifts the minima of the standing wave toward the end. The separation of the first minimum of the standing wave from the surface of the sample is a measure of the dielectric constant.

The original complex transcendental equation derived by Roberts and Von Hippel can be separated into its real and imaginary parts and equating them separately, providing E_{min}/E_{max} is small and that the value of the dissipation factor of the dielectric is less than 0.1. The real parts of the equation are then reduced to:

$$\frac{-\lambda g \tan \frac{360 x_o}{\lambda g}}{2 \pi d} = \frac{\tan \beta_2 d}{\beta_2 d} \qquad (5)$$

where:

λg = Wavelength along the wave guide outside the sample
d = Thickness of the sample
x_o = Distance from the first electric field strength minimum of the standing wave to the face of the sample

From a table of $\tan \theta/\theta$ the value of $\tan \beta_2 d$, $\beta_2 d$ can be readily obtained and the dielectric constant can be calculated from the equation:

$$\epsilon' = \frac{\frac{1}{\lambda c^2} + \frac{(\beta_2 d)^2}{(2 \pi d)^2}}{\frac{1}{\lambda c^2} + \frac{1}{\lambda g^2}} \qquad (6)$$

where:

λc = Free space cut off wavelength

An expression can then be obtained for the attenuation constant by equating the imaginary parts. This reduces to an analytical expression for the total dissipation factor of the wave guide and the sample:

$$\tan \delta_2 = \frac{\Delta x}{d} \left[\frac{\left(\frac{1}{\lambda c^2} + \frac{1}{\lambda g^2} \right) \frac{1}{\lambda c^2 \epsilon'}}{\frac{1}{\lambda c^2} + \frac{1}{\lambda g^2}} \right] \frac{\beta_2 d \left[1 + \tan^2 \left(\frac{360 x_o}{\lambda g} \right) \right]}{\beta_2 d \, (1 + \tan^2 \beta_2 d) - \tan \beta_2 d} \qquad (7)$$

and the dissipation factor of the empty wave guide can be calculated from the following equation.

$$\tan \delta_\epsilon = \frac{\Delta x_\epsilon}{S} \left[\frac{1}{1 + \lambda_c^2/\lambda_\epsilon^2} \right] \qquad (8)$$

where $S = \frac{n \lambda g}{2}$ when $n = 1, 2, 3 \ldots$

and so that

$$\tan \delta = \tan \delta_2 - \tan \delta_\epsilon \qquad (9)$$

Table IV

COMPRESSION TESTS ON FOAMED SILICA SPECIMENS OF VARIOUS DENSITIES

Specimen	Density, lb./cu. ft.	Load, lb.	Psi.
1	45.24	52	250
2	44.93	100	350
3	28.70	97	300
4	41.43	384	700
5	35.07	944	900

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Since approximations are involved in reducing the original complex function to the final expression of dielectric constant and loss tangent, the accuracy of these calculations is about ± 1 per cent, for dissipation factor less than 0.1 and very small E_{min}/E_{max} ratio.

Table III shows the results of these calculations for some of the foamed silica samples.

Figure 6 shows the results of the resistivity versus temperature measurements for sample Si-2.

Figure 7 shows the thermal expansion results for sample Si-8. The sudden decrease in thermal expansion at high temperatures may be due to a slight deformation of the cell structure.

Table IV shows a comparison between density and compression strengths of selected foamed silica samples.

In examining the data and the specimens, it was concluded that the occasional lack of correlation between the microwave properties and the apparent densities of the samples was due to either non-uniform or continuous cell structure, or contamination by minor batch impurities. The volume of bubbles was shown to be directly related to the electrical properties, assuming that the cell structure was uniform, discontinuous, and not contaminated. These are both areas which would bear further investigation should a development program be undertaken.

Applications

A pure foamed silica material would find application wherever good dielectric properties at high frequencies and elevated temperatures are required. Specifically, these materials might be used for insulators in high power radar antenna, or for microwave lens elements such as a Luneberg Lens.

Summary

Several foamed silica samples were evaluated as high temperature substitutes for teflon at microwave frequencies. A 99 per cent pure foamed silica was found to be suitable at temperatures as high as 1100°C.

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Conditions of Glass Formation Among Simple Compounds* IN FOUR PARTS, Part II, Conclusion

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4. Parameters Determining the Activation Energy of Viscous Flow and of Nucleation

a. The Chemical Binding Forces

Our interpretation of the fact that CO_2 at ordinary temperature is a gas whereas the chemically analogous SiO_2 is a three-dimensional polymer is based on the ability of two O^{2-} ions to screen the small carbon core. The core of silicon is larger than that of carbon so that it requires at least four O^{2-} ions in order to be screened. We postulate that the activation energy of viscous flow is the energy which is necessary for the temporary partial unscreening of cations. The anion to cation ratio is one of the most important single parameters which determine the degree of screening and with it the activation energy of viscous flow. We explain the low melting point and the low viscosity of P_2O_5 as compared with SiO_2 on the basis of its higher oxygen to cation ratio. The low boiling point of SO_3 is the result of its high anion to cation ratio which makes it possible for SO_3 to form single molecules like CO_2 at ordinary temperatures. Instead of saying that the bonds in CO_2, SO_3, or P_2O_5 are more covalent than those in SiO_2, we emphasize the extent to which these cations (neon structure) are screened by the number of anions which neutralize their charges.

The ions are not rigid spheres and consequently a simple geometrical picture of the screening of cations and the shape of molecules is not satisfactory. The fact that AgCl has a lower melting point than the corresponding NaCl can be understood on the basis of the mutual electronic interaction. This brings into the picture the polarizing power of the cation and the polarizability of the anion. In spite of the higher lattice energy of AgCl, it melts at a lower temperature than NaCl.

The crystal structure of NaI (NaCl structure) reveals that the Na^+ ion is screened by six I^- ions. The crystal structure of AgI (ZnS - structure) reveals that the Ag^+ ion can be screened by only four I^- ions in spite of its having the same size as the Na^+ ion.

The much lower melting point of B_2O_3 (450°C) as compared with that of Al_2O_3 (2040°C), is the result of the small size and the strong polarizing power of the B^{3+} core (helium configuration) as compared with that of the larger Al^{3+} ion (neon configuration). In the case of B_2O_3 the polarization is more important for the viscosity than even the anion to cation ratio as can be seen by comparing the melting points and the viscosities of B_2O_3 and SiO_2. B_2O_3 melts at 450°C and cristobalite at 1710°C in spite of its higher anion to cation ratio. Fused B_2O_3 has a much lower viscosity than fused SiO_2 at the same temperatures.

We have seen earlier that no valid relation can be found between the bond strength of a compound and its ability to form a glass. We replace the conventional mechanistic concept, namely, that viscous flow and nucleation involve the breakage of existing bonds, by the concept of a temporary partial unscreening of cores. By introducing the polarizability of the ions as a parameter, we abandon the concept of rigid spherical ions and approach the quantum mechanical description of matter as consisting of nuclei which are surrounded by electrons, the density distribution of which depends upon their environment.

Goldschmidt[20] used the bond strength for synthesizing "weakened" and "strengthened" models of a crystal. The bond strength was changed by changing the charge of the ions (valence sum) but retaining their sizes and their ratios. Some properties such as hardness, melting point, and viscosity could be correlated with the strength of the binding forces provided the other parameters remained unchanged. It is reasonable, therefore, to attribute the lower viscosity of BeF_2 as compared with SiO_2 to the different bond strengths. The two crystals and very likely their glasses have similar structures and one can assume that viscous flow involves similar geometrical changes in both cases.

Viscous flow has two aspects; the geometry and the forces. Only if the geometry of flow is the same for two systems can one use the binding forces as the parameter which controls viscosity. We described the activation energy of viscous flow for SiO_2 as the energy needed for temporarily changing some SiO_4 groups into SiO_3 groups or for temporarily moving an O^{2-} ion into a less screening position.

The viscous flow of TiO_2 on this basis might involve the temporary change of some TiO_6 groups into TiO_5 groups. This description based on the change of the coordination number is only a qualitative picture because the screening of the central cation and the degree to which it has to be uncreened during flow cannot be

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described by a number, it depends not only upon the number of nearest anions but also on their distances and their state of polarization. The latter in turn involves their interactions with more distant ions, especially those of the second coordination sphere of the central cation. But this qualitative description is sufficient to show that viscous flow and nucleation of SiO_2 and TiO_2 involves very different geometries. No stable structure is known which contains silicon in threefold coordination. However, in some structures, titanium ions occur in fourfold coordination. The existence of TiO_4 groups in stable structures suggests that the temporary partial unscreening of a TiO_6 group requires less energy than that of a SiO_4 group. This geometrical difference between the two oxides excludes the use of bond strength for deriving or even comparing their activation energies of viscosity and their rates of nucleation.

The activation energy of nucleation is closely related to that of viscous flow; both energy terms depend primarily upon the anion to cation ratio, the polarizability of all ions, and the strength of the binding forces.

Even a heavy ball can be rolled easily on a level surface. A sphere may be called a regular polyhedron with an infinite number of faces. Now let us replace the ball by a regular polyhedron which has a large but finite number of faces and examine what happens to the forces necessary to roll it. Now the rolling motion of the regular polyhedron must involve an upward motion when it moves from one face to the next. As long as the number of faces of the polyhedron is very large so that its shape deviates only slightly from a sphere, the degree to which it has to be lifted during its rolling it also very small. However, if we gradually decrease the number of faces of the regular polyhedron, its shape deviates more strongly from that of a sphere and during its rolling the lifting will require more and more energy. The "ball" now develops an increasing activation energy of rolling. The activation energy of rolling a polyhedron depends of course on its weight, but due to the shape of the polyhedron this relation between activation energy and gravitational forces has become more complex. In the same way the activation energy of ionic reshuffling increases if the coordination of the cation of a substance AX_2 changes from a cube to a octahedron and, finally, to a tetrahedron. Analogous to our rolling polyhedron, the activation energy of flow depends upon the geometry of the system in a way which obscures the relation between binding forces and viscosity and nucleation.

In an earlier paper[3] we described the role which the binding forces play in determining the viscosity of glasses as more "complex" and elaborated on this complexity as follows:

"The fifth important structural parameter is the cooperation of a large number of atoms extending through a considerable volume of glass. This factor is of such importance that one may call it the key to the understanding of mechanical properties of matter on an atomistic basis."

This postulate stands in striking contrast to the conventional treatment of the chemical bond which is assumed to be active between nearest neighbors only. We do not refer here to a modification of existing electrostatic forces by the mutual polarization of ions, but to an effect which can lead to drastic changes of the binding forces within a solid. The cooperation of many atoms in changing the bond strength can be described in different ways and we like to refer to it as a disproportionation of binding forces and of internuclear distances into larger and smaller ones."

This principle accounts for the behavior of glasses under mechanical stress[9] and for the kinetics of melting of simple compounds[15]. It shows why it is not possible to correlate viscous flow, melting, or the mechanical strength of a solid with the average binding forces which are active in the system.

With some exceptions, (the feldspar albite may be given as an example) crystals do not melt because the thermal energy overcomes the average binding forces. The majority of the ionic crystals, e.g., all alkali halides, melt because they develop defects on heating. Each defect, e.g., an anion vacancy, acts as the center of an asymmetry which causes the binding forces of the surrounding ions to disproportionate. In an earlier paper[15] we showed in detail why and how clusters are formed in which the ions are held together by forces which are stronger than the average. At temperatures slightly above the melting point, fused NaCl is fluid because of the weak forces which are active between the individual clusters which oscillate and rotate. The clusters may be called "macromolecules" but one has to remember that the structure of the fused salt is dynamic. The macromolecules do not have long lifetimes and should not be looked upon as permanent structural units.

On the basis of the lattice energy and the thermal expansion of NaCl, one can estimate the average Na-Cl bond strength and the average Na-Cl distance. The high fluidity of fused NaCl, however, cannot be understood on the basis of these values, but reveals that weaker than average forces are active between the ionic clusters. This interpretation of the kinetics of melting focuses our interest on the conditions which favor high concentrations of lattice vacancies in a crystal.

A survey over the chemical compositions of semiconductors reveals that compounds of elements which occur in different valencies due to an incomplete inner shell (elements of the transition groups) can increase their entropy at relatively low temperature by the formation of vacant lattice sites. FeO, NiO or CoO form defective structures which are stable at ordinary or moderately high temperatures, whereas the oxides which contain noble gas-type cations, e.g., MgO, CaO or Al_2O_3, require high temperatures for producing a high concentration of lattice vacancies.

The ability of a substance to increase its entropy by forming a defective structure also increases with increasing polarizability of the anion, i.e., it increases from

the fluoride to the iodide or from the oxide to the selenide and telluride.

On this basis we can understand why oxides of the transition elements have melting characteristics which resemble those of the alkali halides. Heating such an oxide leads to the formation of a large number of defects.

$$V_2^{5+} \ (e^-)_{2x} \ O^{2-}_{5-x} \ (A.V.)_x$$

Vanadium pentoxide assumes the formula indicating that x O^{2-} ions per mole V_2O_5 are missing from the lattice. The oxygen has escaped as elemental oxygen leaving behind x anion vacancies per mole. The 2x electrons of the x O^{2-} ions are quantized with respect to a group of V^{5+} cores, a feature which causes the defective V_2O_5 to develop metallic conductivity. This change lowers the free energy of the system because of the gain of entropy (disorder) due to the presence of vacant anion sites $(A.V.)_x$, and mobile electrons.

We postulate that an oxide of this type will undergo a disproportionation of binding forces on heating and at temperatures above the melting point the liquid will consist of mobile clusters of ions or polymer molecules. Oxides which meet these conditions, e.g., V_2O_5, Nb_2O_5, Ta_2O_5, SnO_2, TiO_2, and TeO_2, do not form stable glasses when pure, even if their geometries would meet any conditions which may be required for the formation of a stable random network. The kinetics of melting and nucleation of these substances is governed by their defects and not by their average bond energies. These crystals can neither be overheated like quartz or albite crystals nor can their melts be easily supercooled.

In contrast to the behavior of V_2O_5 or CoO, the removal of a cation or an anion from a P_2O_5 or a MgO crystal would raise the potential energy of the system much more than can be compensated for by the increased entropy. Hence, MgO develops a high concentration of defects only at very high temperature where the value of TS, i.e., the contribution of the entropy to the free energy, is large. We may explain this difference on the basis that each Mg^{++} ion in MgO requires screening by six O^{2-} ions (MgO has the NaCl structures). Removing some O^{2-} ions and a corresponding number of Mg^{++} ions from the MgO lattice raises the potential energy to a high level because of the high Mg^{++} - Mg^{++} repulsive forces around a vacant anion site. Cations which have a higher polarizability, e.g., Pb^{++} ions, can meet such a situation by distributing their electron density in a way which minimizes the cation-cation repulsion. This is the reason why the oxides of transition elements are more likely to develop defects in the low temperature region than MgO.

Because of their defective structures NiO and CoO turn black at moderate temperatures and develop a considerable electronic conductivity. A solid solution of NiO in MgO remains green when calcined and a solid solution of CoO in MgO remains red. The presence of Mg^{++} ions in the crystals prevents the formation of a major concentration of defects so that these solid solutions have higher melting points than pure NiO and CoO and are insulators. Their colors are characteristic for Ni^{++} and Co^{++} ions in a symmetrical electrical field (aqueous solution).

Later we will return to this stabilization of "perfect" crystals by suitable cations and show how this feature can be used to enforce glass formation in systems which do not contain any oxide which can form a glass by itself.

b. Viscosity Versus Nucleation and Crystallization

Due to the disproportionation of distances and of forces around centers of asymmetry such as anion vacancies, the mobility of particles within a melt is not a simple function of the average bond strength. This lack of a simple relation between the rate of nucleation on one side and thermodynamic and geometrical factors on the other suggests a different approach to the understanding of glass formation. It would be logical to ask the following question: Why can one not bypass factors such as the shape of the molecules, the bond energy of a compound, the field strength of the cations, and the nature of the polyhedra and relate the rate of nucleation and crystallization to a property which integrates over all these factors, namely, the viscosity of the supercooled melt?

Due to thermal fluctuations, an embryo of a crystal will not survive at the liquidus temperature for times which are sufficient for growing into a crystal. A finite degree of supercooling is essential for an embryo of a crystal to form a nucleus which can initiate crystallization. Would it not be reasonable, therefore, to expect that the fluidity of a supercooled melt, say at 0.9 T_L, where T_L is the liquidus temperature in °K, could be chosen as the parameter which controls nucleation and crystallization?

According to this suggestion one would measure the viscosity of the supercooled melt at the temperature 0.9 T_L and use the viscosity or its logarithm as a measure of the ability of the system to form a glass. High viscosity values would mean low rate of nucleation or high probability of glass formation. Viscosities which are below a certain value would indicate that the supercooled liquid has a low probability of glass formation.

One should expect to find a simple relationship between the mobility of the particles as measured by the viscosity of the supercooled melt and the rate of nucleation if the system consists of a random array of identical molecules or of a random three-dimensional continuous network of one kind of polyhedra. These descriptions, however, represent idealized extremes and the real structures of supercooled liquids are likely to differ from both.

We follow G. Hägg's[24] ideas and assume the presence of groups in glasses which are larger than single polyhedra. The atoms in these groups are held together by forces which are stronger than the average forces acting in the system. In this case, both nucleation and viscous

flow involve a partial unscreening of cations, but viscous flow can be possible under conditions in which not all kinds of cations have to be partially unscreened, whereas this may be necessary for nucleation and crystallization.

This situation probably exists in a supercooled melt of anhydrous boric oxide. Such a melt has viscosities which would permit most glasses to devitrify readily. Boric oxide, however, can remain a supercooled liquid for long times. Using Hägg's interpretation, we describe the supercooled melt as consisting of odd shaped groups which are not suitable for assuming a crystalline structure in spite of being able to flow easily.

Another extreme case in which a brittle glass crystallizes is also known. Vitreous arsenic trioxide glass is brittle at room temperature. In spite of its being unable to flow at ordinary temperature, vitreous arsenic trioxide glass devitrifies within a few months and changes into a white porcelain-like mass.

The crystallization of the brittle As_2O_3 glass is a process which is catalyzed by traces of water[25] which lower the activation energy of nucleation. A proton of the water penetrates the O^{2-} ion of the glass thus weakening one As^{3+}-O^{2-}-As^{3+} bond by changing the doubly charged anion into a singly charged OH^- ion. The OH^- ion of the water also enters the same area and participates in the screening of an As^{3+} ion: it lowers the activation energy of nucleation by temporarily and locally increasing the anion to cation ratio. We may describe the catalytic activity of water by the formulas

$$[As_2O_3]_{\text{Glass}} + H_2O \rightarrow [As_2O_2(OH)_2]_{\text{Intermediate}} \rightarrow [As_2O_3]_{\text{Crystal}} + H_2O$$

Both the glass and the crystal have the composition As_2O_3 which means that the water is not used up during the reaction so that it is a true catalyst. The traces of water which the glass adsorbs from the atmosphere open up a new path for the nucleation process by increasing the mobility of the ions within small volume elements, i.e., locally and temporarily. Such a change can aid nucleation but not viscous flow.

We can describe the transformation temperature T_g of a glass as that temperature above which the glass can undergo major structural changes within experimental times. Below T_g we assume that the relaxation times for structural changes which involve the network are too long to be of practical interest, say for its annealing. The temperature T_g can be determined by different methods, e.g., by measuring the thermal expansion. It has been found that T_g is a function of the viscosity of the glass and that for a given method of determining T_g it corresponds to that temperature at which the glass reaches a viscosity value of 10^{13} poises regardless of the glass composition. Such a relation seems to suggest that the relaxation times of structural changes in a glass are controlled by the macroscopic mobility, the viscosity, regardless of the nature of the structural changes which cause the increased thermal expansion. This description, however, is only a useful approximation. Recently O. Lindig[26] proved that no rigorous relation exists between the viscosity and the transformation temperature for different glasses. Using one method and one definition of T_g he found for a group of glasses that this critical temperature varied from 8×10^{11} to 1.1×10^{14} poises.

This variation of the viscosities at T_g over nearly three orders of magnitude shows that the rate of the structural changes which causes the slope of the volume-temperature curve to become steeper is not directly related to the viscosity. We may conclude, therefore, that the viscosity of a supercooled liquid measured at a certain fraction of its absolute liquidus temperature cannot be used as a basis for characterizing its tendency to form a glass or its rate of nucleation and crystallization.

Most changes in the composition of a glass affect its nucleation rate, its viscosity, and its ionic conductivity. All of these properties require the mobility of some particles or processes which involve energy barriers. Frequently one finds that within a limited composition range these properties are affected in a like manner by a composition change. These phenomena, however, involve basically different mechanisms: hence, we cannot generalize and we should not expect a parallelism to exist in the effect of the composition upon the three energy barriers. A certain change in composition might change the energy barriers even in different directions.

5. *Atomic Structure and Glass Formation*

In a criticism of Zachariasen's paper on glass formation Hägg[26] remarked that any approach to the understanding of glass formation ought to start out from the structure of the melt rather than from the structure of the crystal. This is, of course, correct but unfortunately our knowledge of the structures of liquids is still rather scanty and too much has to be left to speculation.

This paper concerns itself with glass formation among simple compounds. An element such as selenium or an oxide such as B_2O_3 represents a system which can be called "simple" from an analytical chemical viewpoint because they contain only one and two kinds of atomic nuclei respectively. This, however, does not mean that a selenium glass or a B_2O_3 glass represents a simple system from a physico-chemical point of view. We attributed glass formation of liquid selenium and liquid sulfur to the anisodimensional shapes of their molecules. The chains are of widely different lengths so that molten sulfur and selenium represent systems of great complexity which consist of a great variety of molecular species.

Glass formation may also result if the liquid contains only one molecular species whose structural configuration (shape) is so odd that it is difficult to establish a self-repeating structure. J. P. Wibaut (1939) reports that glass formation occurs with a number of aliphatic hydrocarbons such as 2-methyl pentane, 3-methyl pentane, 3-

methyl heptane, 3,4-dimethyl hexane and others. The melting points of these compounds are in the neighborhood of—100°C but these odd shaped molecules fail to crystallize and, thus, solidify as glasses. The forces which act between hydrocarbons are primarily of the van der Waals' type, i.e., they decrease very rapidly with increasing distance. Considering the odd shapes of these molecules it might well be possible that a condensation to a non-crystalline solid might lower the free energy more than one which has periodicity but requires a more spacious arrangement. It seems doubtful, however, that this kind of glass formation has its analogues in the realm of inorganic compounds.

H. Salmang[28] in a recent publication reminds us of the importance of the molecular concept which was used for interpreting the structure of glass long before the concept of the continuous network was proposed. A good part of our fundamental knowledge in nucleation stems from G. Tammann's[29] work on glasses which, like salol and other organic substances, consist of molecules. The development of the network concept seemed to have made the molecular approach obsolete. B. E. Warren[30] and his school could prove that silica molecules did not exist as individual units, either in crystals or in glasses.

The molecular concept, however, is still important because some substances, e.g., elemental sulfur and the oxides of arsenic and phosphorus, can form both types of solids, one which consists of molecules and another whose structure is continuous in one (chains), two (layers), or three directions. Substances of this type show anomalous physico-chemical properties, e.g., within a certain temperature region their vapor pressures may decrease on heating and their viscosities may increase.

Experimentally one determines the ability of a compound to form a glass by heating it above its melting point and allowing the melt to cool at a certain rate. By this method we can establish the fact that silica is a glass-forming oxide. We take it for granted that this property is characteristic for the chemical substance silicon dioxide and not just for one of its modifications, e.g., for quartz, tridymite, or cristobalite. As long as the temperature is sufficiently high to melt the crystal completely, one obtains a glass on cooling.

This, however, is not the case for sulfur. Elemental sulfur melts at 119°C, but heating it to 120°C produces a melt which does not form a glass, no matter how fast it is cooled. In order to produce amorphous sulfur one has to heat the melt to a higher temperature. Beginning at 150°C the S_8 molecules (which are the constituents of both the crystalline sulfur and the liquid) which are stable a few degrees above the melting point, break open and the S_8 rings form chains which may become very long. G. Gee et al.[31,32] found that chains are formed which contain up to 10^6 atoms. The formation of these macromolecules from the S_8 rings causes the viscosity of the melt to increase a thousand times if its temperature is raised from 157° to 187°C. In this temperature region equilibria are established fast between molecules of widely different molecular weight and chain lengths.

The oxides P_2O_5 and As_2O_3 behave like elemental sulfur. The crystalline P_2O_5 consists of P_4O_{10} molecules and so does its vapor. The vapor pressure of P_2O_5 decreases and the viscosity of the molten P_2O_5 increases if the temperature is raised further. The atoms in the individual molecules reorganize and form a variety of macromolecules, most likely chain-like polymers analogous to molten sulfur.

We are accustomed to think of glass formation being enhanced by lowering the temperature of a system rapidly, and nucleation and crystallization being enhanced by slow cooling rates. This, however, may not apply to systems which contain different molecular species. H. Bötticher et al.[33] (1951) found that As_2O_3 vapor chilled from 400°C to room temperature produced a crystalline modification of As_2O_3 (arsenolite), whereas slow cooling caused a condensation of vitreous As_2O_3 at 315°C. X-ray examination of the As_2O_3 glass revealed that its close-range order resembles that of claudetite more than that of arsenolite. Arsenolite is built of As_4O_6 molecules, the same molecules which are present in the vapor phase. Claudetite is a two dimensional polymer, it forms a layer lattice and the layers are formed from infinite arrays of ions.

From the viewpoint of the glass technologist the complexity of fused sulfur, selenium, phosphorus pentoxide and arsenic trioxide is of little practical interest so that these glasses have not received much attention lately. However, we may find that a similar complexity of the melt is responsible for the behavior of one of our most important glass-forming oxides, namely boric oxide. How does the behavior of boric oxide compare with that of vitreous sulfur?

We have no final picture yet of the constitution of B_2O_3 glass or of the structure of fused B_2O_3, but we must assume that the structure is one of great complexity. It is not our object to discuss here the constitution of glasses but we have to recall a few facts which are pertinent to glass formation. Fused B_2O_3 is very difficult to crystallize in the pure state at ordinary pressure. L. McCulloch[34] (1937) obtained pure crystalline B_2O_3 accidentally from a melt which contained some water. We must assume that the stability of a B_2O_3 glass or the reluctance of the melt to crystallize even on very slow cooling is a matter of its complexity rather than of its viscosity.

The activation energy of nucleation of simple compounds is related to the activation energy of viscous flow if the latter represents the highest energy barrier which obstructs the path of the transition of the supercooled liquid into the crystal. In vitreous elemental sulfur, we expect to find two energy barriers, one which affects the viscosity of the melt and another which affects the formation of S_8 ring molecules from the long sulfur chains. For elemental sulfur, the formation of S_8 ring molecules seems to have a low energy barrier because

even at ordinary temperature plastic sulfur changes into a crystalline modification within a few days.

One may assume that the supercooled melt of sulfur consists of polymers which have very different molecular weights. One may also assume that the distribution function of the molecular weight in such a system represents an equilibrium which is temperature dependent. At any given temperature a finite number of chains exists which contain eight sulfur atoms. This molecular species is the only one which can form a stable ring and S_8 rings are the only particles which can aggregate and build up a sulfur crystal at temperatures below the melting point.

It is a characteristic feature of molten sulfur that the internal equilibrium is established rapidly. If we were able to remove S_8 molecules from the melt their concentration would not be lowered materially because they would reform from other polymers in order to retain that distribution function which has the lowest free energy. If the activation energy of S_8 ring formation were high as compared with kT, the supercooled sulfur either would have to crystallize in a fibrous modification by an orientation of its chains analogous to selenium or it would have to remain a glass.

The most characteristic difference between glasses and crystals is the strong dependence of all properties of glasses on their previous thermal history. The establishment of internal equilibria in glasses requires a period of time which is sufficiently long to permit the freezing in of a high temperature structure. All glasses, therefore, have densities which may vary within a certain range depending upon the rate at which they have been cooled. Vitreous boric oxide, however, shows this phenomenon to an unusually high degree. G. W. Morey[35] discussed widely different values of the density of vitreous B_2O_3 which can be found in the literature and he remarked that this glass is "extraordinarily susceptible to heat treatment." He reports densities which vary from 1.778 to 1.838 depending on the rates of cooling.

Vitreous boric oxide also shows an unusually strong change of its thermal expansion when heated above its softening temperature (T_g). As a rule the coefficient of thermal expansion of a glass is lower below T_g than above. Vitreous B_2O_3 follows this rule, but the difference between the temperature coefficients of thermal expansions below and above T_g is unusually large. According to A. Cousen and W.E.S. Turner[36] (1928), a B_2O_3 glass expands its volume nearly ten times as much above the softening range as below it.

K. Fajans and S.W. Barber[11] (1952) reached the conclusion that fused B_2O_3 on cooling undergoes a structural change which weakens the structure. A system which retains its geometry on heating should have a viscosity which decreases with increasing temperature according to an exponential function. Indeed, plotting the logarithm of the viscosity of fused B_2O_3 versus 1/T one finds a linear relationship within the temperature range of 220-370°C. In this temperature region the activation energy of viscous flow is constant. At higher temperatures, however, the viscosity of B_2O_3 is higher than one would expect from an extrapolation of its low temperature viscosity. This increase of the activation energy of viscous flow with the temperature is interpreted by Fajans and Barber as indicating the formation of B_4O_6 molecules at low temperature. These molecules, like the S_8 molecules in fused sulfur, change into larger particles on heating which lead to a stronger structure at higher temperature. On this basis Fajans and Barber explain the fact that the viscosity of B_2O_3 at 727°C is more than 6000 times higher than the value which one obtains by extrapolating the low temperature viscosity to this temperature.

J. Goubeau and H. Keller[37] (1953) interpret the Raman spectrum of vitreous B_2O_3 as indicating boroxol groups, i.e., six membered rings of BO_4 tetrahedra. These rings have been found in some crystalline borates.

V.V. Tarasow and E.F. Stroganov[38] (1956) interpret the low temperature heat capacity curve of vitreous B_2O_3 as indicating chain structures.

Recently, J. Krogh-Moe[39] presented a critical discussion of the earlier views and his own views on the structure of vitreous B_2O_3 and some borate glasses. His interpretation which is based on X-ray diffraction and on infrared spectra of various boron compounds is centered around the changes of the coordination of boron and the asymmetry of the structural units.

Up to the present time all scientists have attempted to describe the structure of a B_2O_3 glass as a random three-dimensional network which is continuous, inasmuch as it consists of polyhedra which are interlinked by sharing O^{2-} ions so that infinitely extending sheet structures result. It is more likely that G. Hagg's description is far better, i.e., that vitreous B_2O_3 consists of molecules which have widely different molecular weights and odd shapes.

The different views on the structure of vitreous B_2O_3 must not necessarily be controversial. There is no reason why the simultaneous presence of B_4O_6 molecules, boroxol rings, and BO_4 chains should be ruled out.

The density of crystalline B_2O_3 has been determined by S.V. Berger[40] (1953) to be 2.56 which is much greater than even the highest value of the density of vitreous B_2O_3 (1.86) as reported by L. Shartsis, W. Capps and J. Spinner[41] (1953). The entropy of fusion of B_2O_3 (7.3 e.u.) is considerably higher than that of silica (0.9 e.u.).

These facts suggest that the structure of vitreous boric oxide is very complex and is not likely to resemble that of the crystal. Fused boric oxide has a transformation temperature below 250°C and a melting temperature of 450°C. Its unusually low rates of nucleation and crystallization cannot be explained on the basis of viscosity alone. We assume, therefore, that the activation energy of nucleaton is determined by the change of the structural units into those which are suitable to form a crystal. This change may involve B_4O_6 molecules, as

well as rings, chains or two dimensional layers of polyhedra. The crystallization of vitreous B_2O_3 has as a prerequisite the change of existing building units into different structural units which may have a different short range order. Viscous flow is possible without such a structural change but nucleation is not.

According to our concept the crystallization of B_2O_3 from the glass is a process which is analogous to that of plastic sulfur only inasmuch as it involves a change of the existing structural elements into others which make it possible to establish long range order. It differs from that of sulfur inasmuch as the activation energy of the structural change for B_2O_3 is high as compared with that of viscous flow.

Earlier[15] we discussed similar structural changes in alumino-silicates. The lack of nucleation of an albite glass and the inability of such a glass to completely crystallize when seeded by albite crystals are phenomena which are due to the need of formation of structural units in the supercooled melt which are different from those which participate in the rapidly cooled glass.

6. *Effect of Minor Additions on Nucleation*

We attribute the high viscosity of fused silica to its close resemblance to a "perfect" cristobalite crystal, in particular, to the absence of a major concentration of vacant anion and cation sites. Even at the high temperature which is needed in order to melt SiO_2, the silicon cores remain screened by four oxygens. The activation energy of viscous flow is high because flow requires a temporary partial unscreening of Si^{4+} cores. The rigid screening demand of the Si^{4+} cores makes it impossible for the structures of cristobalite and of vitreous silica to increase their entropies by forming a high concentration of vacant anion sites.

A. Dietzel and H. Wickert[42] examined the system SiO_2-Na_2O with respect to the stability of the supercooled state or the tendency of a melt to form a glass on cooling. In order to describe the tendency of a melt to solidify as a glass quantitatively, they use the reciprocal value of the maximum rate of crystallization which they called "Glasigkeit" (glassiness). According to Dietzel and Wickert the addition of alkali to silica does not change the glassiness in a monotonic function but its values goes through maxima and minima. The rate of crystallization of sodium metasilicate is fastest for a glass which has the composition of the metasilicate. Similarly the rate of crystallization of the sodium disilicate is fastest for a sodium disilicate glass. Minima of the values for the glassiness, i.e., maxima for the rates of crystallization, are found for glasses whose compositions are the same as the crystals which precipitate on cooling. This relation seems to be very plausible and it is exactly the behavior one would expect. On the same basis one might also expect pure silica glass to have a maximum rate of crystallization just as glasses which have the compositions corresponding to the crystalline phases $Na_2O \cdot SiO_2$ and $Na_2 \cdot 2SiO_2$ show a maximum rate of crystallization. This however, is not the case. Pure vitreous silica shows little tendency to crystallize: its glassiness is very high. However, the addition of Na_2O enhances crystallization to such an extent that Dietzel and Wickert had to develop special melting methods for producing small quantities of glasses in the composition range 0.5 per cent—12 per cent Na_2O.

This effect of the addition of Na_2O on the nucleation of cristobalite calls for an explanation because it is just the opposite of what might be expected for two reasons:

1. The addition of Na_2O to SiO_2 lowers the liquidus temperature and, thus, decreases the kinetic energy which is available for nucleation.
2. The addition of Na_2O to silica changes the glass composition in a direction away from the phase (cristobalite) which crystallizes.

a. Positive Nucleation Catalysts

We learned that the presence of traces of water at the surface of vitreous arsenic trioxide makes it possible for this glass to crystallize in its brittle state. In a similar fashion, alkali acts as a catalyst for the nucleation and, generally speaking, for phase transformations of silica.

The presence of alkali increases the screening of the Si^{4+} cores by increasing the anion to cation ratio and by introducing O^{2-} ions of greater polarizability than those present in pure silica. Alkali catalyzes the rearrangement of the SiO_4 tetrahedra because it introduces into the structure an asymmetry and it provides additional screeners. Alkali is a catalyst for the transformation of quartz into cristobalite. The experiments bear out the fact that this catalytic action of Na_2O must be much more important for the rate of nucleation of cristobalite than the two antagonistic effects listed above.

Recently S.D. Brown and S.S. Kistler[43] (1959) studied the devitrification of some high SiO_2 glasses in the system SiO_2-Al_2O_3 and found that the growth rates for all specimens with added Al_2O_3 are greater than those of pure silica. They wrote: "Inasmuch as the same crystalline phase (cristobalite) separated in all cases and inasmuch as the liquidus temperature of the Al_2O_3-containing glasses are less than those of pure SiO_2, one might expect results just the opposite from those here presented."

These observations show that even Al_2O_3 is a positive catalyst. It probably participates in the structure of silica as AlO_4 and AlO_6 groups. Its much weaker effect upon nucleation rates as compared with Na_2O is no doubt concerned with the much lower polarizability of the O^{2-} ions which Al_2O_3 introduces into the system. The asymmetry introduced by alumina in silica is certainly less than that introduced by alkali.

The catalytic activity of small percentages of H_2O and HF are well known to the geologist. He associates their presence in a magma with its high rate of crystallization. A magma which contains a sufficient concen-

tration of these "mineralizers" forms a crystalline rock, whereas the so-called dry magma fails to crystallize on cooling and forms obsidian.

These mineralizers raise the anion to cation ratio of the system, thus introducing additional screeners. OH^- ions and F^- ions which take the place of a regular O^{2-} ion are centers of an asymmetry which lead to a disproportionation of binding forces in a fashion similar to that produced by an anion vacancy.

b. Negative Nucleation Catalysts

Negative catalysts are substances which slow down a reaction rate, not necessarily by increasing the energy barrier but most frequently by paralyzing the influence of a positive catalyst which is present in the system. We saw that Na_2O is a very strong positive catalyst for the nucleation of cristobalite and Al_2O_3 is a weak positive catalyst. However, introduced into a sodium silicate glass Al_2O_3 becomes a network former which counteracts the effect of Na_2O by eliminating the single bonded O^{2-} ions. B_2O_3 has a very similar effect. These oxides are widely used in technical soda lime silicate glasses for suppressing devitrification and for increasing the chemical resistivity. In a soda lime silicate glass both effects go parallel and both are based upon the same structural change. Alumina and boric oxide immobilize the O^{2-} ions which the alkali introduces and they lower the polarizability and the screening power of the O^{2-} ions, thus increasing the symmetry of the structure.

The immobilizing action which alumina exerts upon the single bonded O^{2-} ions of a sodium silicate glass reveals itself in the internal friction or the anelasticity. L. C. Hoffman and W. A. Weyl[44] showed that the partial replacement of SiO_2 by Al_2O_3 in a series of sodium magnesium silicate glasses of the general formula Na_2O, MgO, x Al_2O_3 (4-x) SiO_2 had little effect on the maximum of the internal friction at 0°C, whereas at 100°C it lowered the internal friction drastically. H. Rötger[45] attributes the low temperature peak to the mobility of the Na^+ ion and its response to the mechanical deformation of the glass. The next higher peak can be attributed to the most polarizable O^{2-} ions which are present in the glass. Nucleation cannot be aided by the mobile Na^+ ions but only mobile better screening O^{2-} ions. This explains why the presence of alumina decreases the rate of nucleation as well as that of corrosion in glasses of this and similar compositions without decreasing the mobility of the alkali ions.

IV. Conclusions and Summary

Glasses are amorphous solids which have formed from the liquid state on cooling in a reversible process. Hence the one fundamental condition which a substance has to meet in order to be able to form a glass is that it is liquid or that it can be changed into a liquid. Most substances which can be melted, e.g., all the true metals and all alkali halides, crystallize on cooling. Glass formation is the result of either one or a combination of the following two conditions.

1. The substance melts as a "perfect" crystal, i.e., on heating the kinetic energy or the thermal agitation gradually overcomes the lattice forces. The crystal at its melting point does not have a major concentration of defects. Substances of this type, e.g., quartz, have sharp melting points only in the thermodynamic but not in the kinetic sense.
2. The substance melts and changes at the melting point or at a higher temperature into a system which represents a mixture containing a large variety of structural units, e.g., rings, chains of different lengths, etc. In this case glass formation is possible because the complexity of the melt lowers the probability of successful collisions of any one particular species which might produce a crystal.

These two types of glass formation in their purest form are represented by silica on the one side and elemental sulfur or phosphorus pentoxide on the other. These compounds represent the extremes, but one finds all degrees of combinations of the two types to be effective in glass formation.

Albite is a typical example for the combination of the two extremes. Albite melts as a perfect crystal which can be easily overheated for long periods of time. Its melting point (1118°C) can be determined only by static but not by dynamic methods. At a temperature which is higher than its melting point (1600°C), the melt still has the chemical composition of albite but it no longer contains those structural elements which were present in albite and which are essential for forming an albite crystal from the glass: new structural units are formed. Whereas the establishment of the internal equilibrium between the molecular species is relatively fast in sulfur, a long time is required for albite so that, in contrast to sulfur, a melt of albite forms a very stable glass even when it is cooled very slowly.

Use of the second condition which leads to glass formation, namely, the higher complexity of the melt, is widely practiced in the manufacture of glasses, glazes and enamels. Most technical glasses have complex compositions. This type of glass formation, however, is not limited to complexity with respect to chemical composition but applies equally well to complexity from a physical-chemical point of view. Hence, simple compounds such as sulfur and P_2O_5 can form glasses on the basis of the structural complexity of their melts. In their crystalline state both substances contain well defined molecules, namely, S_8 and P_4O_{10} molecules. At temperatures above the melting point these small molecules change into macromolecules: chains of widely different lengths. It is the great variety of the molecular species which introduces complexity into these simple

(*Continued on page* 658)

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Bibliography on Glass Structure*

PART V

129. K. S. Evstrop'ev

"Refraction of the Oxygen Ion in Silicate and Borate Glasses," J. Phys. Chem. U.S.S.R. 20, 605-12 (1946).

The refraction due to O, calculated from literature data, is different for SiO_2 and B_2O_3 glasses and depends on the energy of the lattice. The separation of the oxygen ions in glasses into "structure ions" and "anions" often conflicts with other data on the polarizability of ions.

130. K. S. Evstrop'ev

"Refraction of the Oxygen Ion in Silicate and Borate Glasses, I," J. Phys. Chem. U.S.S.R., 20, 561-6 (1946).

Refractivity of the oxygen ion in glasses is discussed.

131. R. H. Thomas

"Atomic Structure of Glass," Ceram. Age, 50, 115 (1947).

A compilation and bibliography of the variable literature on x-ray studies of vitreous structures.

132. Kuan-Han Sun

"Fundamental Conditions for Glass Formation," J. Soc. Glass Tech., 31, 245-53 (1947).

The fundamental condition for glass formation is the existence of strongly bonded large networks or long chains of atoms in the liquid. Such chains or networks will not only prevent or hinder the orientation or arrangement of atoms in the chains or networks themselves but also that of the other atoms not in the networks in the liquid near the melting point. Thus, a liquid structure will persist even after the liquid has been cooled below the melting point or liquidus temperature and indeed farther down to such temperatures that free long range movement of atoms becomes impossible. In order to obtain such long chains or large networks it is necessary that the bond strength of atoms in the chains or networks be very strong, small ring formation of these strongly bonded atoms be at a minimum, and the relative numbers of various atoms in the chains be such that a chain is structurally possible.

133. W. Hurd Safford and Alexander Silverman

"Alumina-Silica Relationship in Glass," J. Am. Ceram. Soc., 30, 203-11 (1947).

Structure and coordination numbers of Al and Si are worked out on the basis of molar refraction measurements.

134. Taro Moriya

"Structure of Glass," J. Japan. Cer. A., 55, 60-62 (1947).

Moriya describes his theory on the structure of glass. Glass consists of hundreds of high molecular cells and many ions and molecules which surround the cells. This combination of high molecular cells has no regularities like crystals and the cells are quite amorphous or quasi-crystalline. The cells have definite chemical compositions. When fused glass cools the cells grow and their number increases with the loss of surrounding simple molecules.

135. B. Long

"The Vitreous State," Verre, 2, 1-10 (1947).

This review of the constitutional glass problems stressing the crystallite hypothesis at a time when it is unfashionable now proves interesting.

136. R. W. Douglas

"The Relation Between Physical Properties and Structure of Glass, I, Variations of the Physical Properties with Time," J. Soc. Glass Tech., 31, 50-73 (1947).

The x-ray diffraction patterns of liquids and glasses are compared; it is shown that the similarities between them enable a qualitative application to be made to glasses of some theoretical work on the liquid state by J. D. Bernal. In this way a satisfactory account may be given of the time variation of the physical properties of glasses in the temperature range in which their viscosity increases above 10^{13} poises. This account is given in terms of a random network structure and involves no considerations of aggregation or association.

137. H. Cole

"Glass-Network Formation and the Covalency Factor," J. Soc. Glass Tech., 31, 114-21 (1947).

The various expressions which have been derived to determine whether an element acts as a network former in glass or as a stabilizer or modifier are discussed. On the basis of the author's theory of glass structure, in which such atoms as sodium are considered as playing a more or less important part in the maintenance of the continuous network, a covalency factor is assigned to various elements. It is emphasized that very strong bonds tend to produce regular crystalline

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atomic arrangements, and that normally weak bonds may be constrained by the rest of the structure to have some additional covalent character. The arbitrary classification of elements into glass formers, stabilizers or intermediates, and modifiers cannot be supported.

138. H. Cole

"The Constitution of Colored Glasses," J. Soc. Glass Tech., 31, 105-113 (1947).

Coloring agents in glass are classified on the basis of the analogy between glasses and solutions.

139. H. Cole

"The Color and Constitution of Glass," J. Soc. Glass Tech., 31, 100-4 (1947).

The bonding of the atoms in glass is considered from the point of view of the electronic theory of valency.

140. H. Cole

"Constitution of Alkali Halide-Borate Glasses," Nature, 160, 191 (1947).

Cole disagrees with Majumdar et. al. on the fact that their otherwise correct data substantiates the arrangement postulated for chlorides in borate glasses.

141. Joseph S. Lukesh

"Application of Crystal Chemical Principles to the Problem of Glass Structure," Crystallographic Soc. Annapolis, Md., March 1947; Cer. Abs. 69_g (1948).

142. Kuan-Han Sun

"Fundamental Conditions of Glass Formation," J. Am. Ceram. Soc., 30, 277-81 (1947).

143. W. A. Weyl

"The Surface Properties of Glasses as Effected by Heavy Metal Ions," J. Soc. Glass Tech., 32, 247-59 (1948).

This paper provides a common theoretical basis for subsequent publications on the hygroscopicity of glass, adhesion phenomena and on the catalytic action of glass in respect to the gas nucleation and thermal decomposition of metastable compounds. Four new methods are discussed which provide better insight into the structure of the glass surface at room as well as at higher temperatures.

144. J. M. Stevels

"The Properties of Optical Glass in Connection with its Structure," Verres et Refrac., 4-14, Feb. 1948.

The absorption and structure of glass are correlated.

145. J. E. Stanworth

"The Transformation Range in Glass," J. Soc. Glass Tech., 32, 301-8 (1948).

The transformation range is discussed with particular reference to glycerol and boric-oxide glasses.

146. J. E. Stanworth

"On the Structure of Glass," J. Soc. Glass Tech., 32, 154-72 (1948).

The paper discusses certain criteria for glass formation in pure oxide and complex oxide glasses, and stresses the importance for the glass-forming cation of a low coordination number and ability to form covalent bonds to oxygens.

147. J. E. Stanworth

"The Viscosity and Nature of Glass," J. Soc. Glass Tech., 32, 20-31 (1948).

The viscosity-temperature properties of glass are discussed on the basis of the theories of Frenkel and Eyring, and it is concluded, in contrast to the views of Seddon, that the moving particles, at least in the temperature range above the annealing temperature, are not large compared with single molecule size.

148. A. Smekal

"Present and Future Position of the Physics of the Glassy State," Glastech. Ber., 22, 35 (1948).

Glasses were formed, in general, by the solidification of supercooled liquids in the absence of crystal growth nuclei, also by polymerization and multicondensation. Vitreous amorphous states, however, could also be obtained by special methods of separation, such as, for example, electrolysis. Systematic investigation into the range of variability of the most important properties and into the limits of stability of these glass states still remains to be done.

149. Hassan-Ali Sheybany

"Structure of Mixed Alkali-Silicate Glasses," Verres et Refrac., 2, 127-45, 229-42, 363-75 (1948); 3, 27-39 (1949).

Physical properties, maxima and minima are explained on the basis of electronic structures, polarization and packing effect.

150. M. K. Roman, E. C. Marboe and W. A. Weyl

"The Influence of Heavy Metal Ions on the Tygroscopicity of a Glass Surface," J. Soc. Glass Tech., 32, 260-63 (1948).

The affinity of a glass surface to water as affected by the surface ions is discussed.

151. Anenta Winter-Klein

"Recent Progress in Research on the Structure of Glass," Rev. optique, 27, 249-57 (1948).

X-ray studies indicate that the regular grating built up of elementary SiO_4 tetrahedrons in crystalline quartz is replaced in glass by a disordered arrangement when distances are considered that are large compared with interatomic separations. Similar considerations apply to glass containing B_2O_3 and P_2O_5. If Na_2O or CaO is added to SiO_2 glass, all the oxygen atoms possible enter into elementary SiO_4 groups containing a grating with lacunas into which the Na or Ca atoms go. Such structure accounts for the anomalous coefficient of expansion of B_2O_3 glasses, explains why glass has no fusion point, and explains the role of CaO or Na_2O in activating the melting of a mixture of these oxides with silica. In a glass of composition 6 $SiO_2 \cdot CaO \cdot Na_2O$ there is 92% oxygen by volume and this shows why 95% of the refractivity of glass is due to oxygen.

152. I. Ackermann

"The Structure of Glass," Optik 3, 47-52 (1948).

Electron diffraction patterns of silica glass films are similar to those obtained for a glass or liquids. Although the "distance d of lattice planes" in glass,

completed from the broad inference rings, are practically the same as those computed from the sharp rings for the interplanar distances in cristobalite, the close agreement does not allow a decision between the two possibilities for the glass state viz. a mixture of tiny separate crystallite or a continuous irregular lattice pattern.

153. N. J. Kreidel

"Problems Relating to Glass Structure," Ceram. Age, 52, 255-8, 275-6 (1948).

Kreidel discusses the structure of complex glasses and limitations of present knowledge which prevents one from forming a simple concept of structure.

154. A. G. Smekal

"A Note on Some Relations Between the Mechanical Strength and the Structure of Glass," J. Soc. Glass Tech., 32, 378-81 (1948).

A resume of work carried out during the recent years in Germany.

155. J. E. Stanworth

"The Ionic Structure of Glass," J. Soc. Glass Tech., 32, T366-72 (1948); cf. C. A. 43, 2837e (1949).

Arguments are presented in favor of regarding glasses as collections of ions arranged in space in a matter having general similarity to that observed in crystals though with characteristic differences.

156. Henry H. Blau and James A. Johnson

"Investigations of the Glass Structure Using Radioactive Tracers," Ohio State Univ. Eng. Exp. Station News 20 (5) 45-47 (1948); Cer. Abs. 142g (1949).

157. H. O'Daniel

"Structure and X-ray Investigation of Glass," Glastech. Ber. 22, 11-17 (1948).

This reports of the very elaborate discussion on the principle theories which combine the structures of liquids with that of glasses, and of the application of crystal chemical methods and rules to glass constitution. The analogy of silicate, borate, phosphate glasses with organic and "semi-organic" high polymeric glasses is discussed with a particular emphasis given to the distinction of ionic and interatomic forces of bonding. The difficulties of the x-ray investigation of glass are illustrated, and the necessity of combining the theory with better known details of crystal structures. Warren's and Zachariasen's fundamental ideas on the random network structures must in future investigations be combined with modern ideas of the molecular constitution. The study of organic and "semi-organic" high polymers will probably show the most successful way to this end.

158. J. M. Stevels

"Progress in the Theory of the Physical Properties of Glass," Monograph Progress Research in Holland, No. 20, 104p (1948); cf. C. A. 42, 6501gh (1948).

159. H. O'Daniel

"Concerning the Structure and X-ray Study of Silicate Glasses," Glastech. Ber. 22, 11-15 (1948).

A brief review of the results obtained in the study of crystals and glasses and in particular of silicates and glasses, by x-ray diffraction techniques. It was probable that Warren's interpretation of the results in terms of a three dimensional random network did not represent the whole truth. It seemed more likely that silicate glasses should contain similar structural groups to those which occurred in silicate crystals. Thus, it is suggested that a silicate glass consisted of a random arrangement of one, two and three dimensional groupings, together with small isolated rings of tetrahedra.

160. Walter Kauzmann

"The Nature of the Glassy State and the Behavior of Liquids at Low Temperatures," Chem. Revs. 43, 219-56 (1948).

A review.

161. M. Fanderlik

"What We Know About the Structure of Glass," Sklarsk Rozhl, 24, 1-4 (1948).

A review.

162. H. Ebert

"Glassy State," Glashutte 75, 105-6 (1948).

The glassy state is characterized, according to thermodynamic or continuum theory, by the transformation point, which is the intersection of two lines representing the thermal expansions below and above this point and by structural or discontinuum theory.

163. A. Dietzel

"The Structure and Properties of Glass," Glastech. Ber., 22, 41-50 (1948); 81-86 (1948); 212-24 (1948).

A complete treatise on glass structure including introduction, properties of ions, conditions of compound formation, structures of unitary glasses, structures of binary glasses, structures of polynary glass systems, foreign anions, coloration and appendix.

164. George S. Bachman

"Infrared Study of the Structure of Glasses Prepared by Melting Crystals," GLASS INDUSTRY 29 (8) 435-37, 466 (1948).

The infrared transmission spectra of powdered silicate glasses prepared by melting crystals are examined in an attempt to cast some light on the structure of glasses. It is concluded that glasses of the same composition may differ somewhat in internal structure, that there is a relationship between the structure of the glass and that of the parent crystal, if the temperature at which the crystal is melted to form the glass it not too high and that when the temperature is high enough to cause sufficient mobility of the glass, this structural relationship between crystal and glass gradually disappears.

165.

"Some Aspects of the Nature and Constitution of Glass," Nature (4128) 162, 938-39 (1948).

Proceedings of the Symposium on Some Aspects of the Nature and Constitution of Glass and the joint meeting of the International Commission on Glass and the Society of Glass Technology, Sept. 30, 1948.

166. R. W. Douglas and G. A. Jones

"A Study of Configuration Changes in Glass by Means of Density Measurements," J. Soc. Glass Tech., 32, 309-39 (1948).

167. W. A. Weyl

"Surface Structure and Surface Properties of Crystals and Glasses," J. Am. Ceram. Soc., 32, 367-74 (1949).

The difference between the surface structure and the interior of a crystal depends on the polarization properties of the ions. A schematic representation of the bond strength distribution in ideal and real crystals, in glasses and in surface films brings out the similarity between the surface of crystals and the structure of glasses.

168. W. A. Weyl

"The Dielectric Properties of Glass and Their Structural Interpretation," J. Soc. Glass Tech., 33, 220-38 (1949).

Our knowledge of the atomic structure of simple glasses has been advanced sufficiently to be used as a basis for interpreting their properties. The dielectric properties of glasses are determined by electronic and atomic polarization processes which are not different from those encountered in crystalline solids.

169. J. M. Stevels

"Relations Between the Structures of Glasses and Their Mechanical, Physical and Chemical Properties," Verres et Refrac., 3, 359-68 (1949); cf. C. A. 42, 6501gh (1948); cf. Zachariasen, C. A. 26, 5800 (1932).

The author discusses Smekal's new theory of mixed bonds and his own contributions, such as the extremes in property-composition relations. Various examples of minima and saddle points in dielectric losses in glasses as a function of binary variations of a pair of components are given.

170. A. Smekal

"The Nature of Glass Forming Substances," Glastech. Ber., 22, 278-89 (1949).

No glasses are formed by compounds representing one of the four principal types of bonds: ionic, atomic, intermolecular, or metallic. Glasses are formed characteristically by substances showing mixed bonds in suitable relations.

171. Alexis G. Pincus

"Influence of Structural Chemistry on Glass Composition," Ceram. Age, 54 (5), 299-306 (1949); Cer. Abs. 67j (1950).

Just as crystal chemistry has brought order to the classification of silica minerals and an understanding of the complexity of their compositions, it has also helped to organize and extend the range of compositions that can be obtained as glasses. Nineteenth century work on glass compositions, early Twentieth century glass, new optical glasses of the Eastman Kodak Company, other changes in glass compositions, and future fundamental researches on glass are discussed.

172. G. O. Jones and F. E. Simon

"What is Glass," Endeavour, 8, 175-81 (1949).

The properties of glass are discussed in thermodynamic terms, by treating glass as a nonthermodynamic phase.

173. Frank L. Jones and Norbert J. Kreidl

"Optical Properties and the Constitution of Glass," J. Soc. Glass Tech., 33, 239-54 (1949).

The classification of constituents as network-formers, modifiers or intermediates must be revised in certain glass-forming systems. The function of constituents is influenced not only by their charge and apparent radius, but also by the packing and the electron distribution in the structure.

174. A. Danzin

"Actual Knowledge About the Structure of Mineral Glasses," Annales de Radio electricite, IV, 249-56 (1949).

A critical review of published theories on glass constitution which basically favors the Zachariasen-Warren theory in spite of the "danger of the mental satisfaction it provides."

175. H. J. Arnikar

"A Note on the Constitution of Glass: The Degree of Alternation of the Constituent Oxides," Trans. Indian Ceram. Soc., 8, 31-8 (1949).

176. A. Pevny

"The Structure and Properties of Glass," Chem. Zvesti, 3, 106-15 (1949).

177. E. Kordes and H. Becker

"Physiochemical Investigation on the Fine Structure of Glasses, V, Glasses of the Binary System of Phosphorous Pentoxide with Cadmium, Sodium and Lithium Oxides," Z. anorg. chem., 260, 185-287 (1949); cf. C. A. 36, 1232[7] (1942).

Previous work of Kordes is extended to these binary systems. Methods for the preparation of the glasses as well as density, molar volumes, n.s., and molar refractions are given.

178. G. Ruess

"Raman Spectra and Glass Structure," Glastech. Ber., 22, 173-7 (1949).

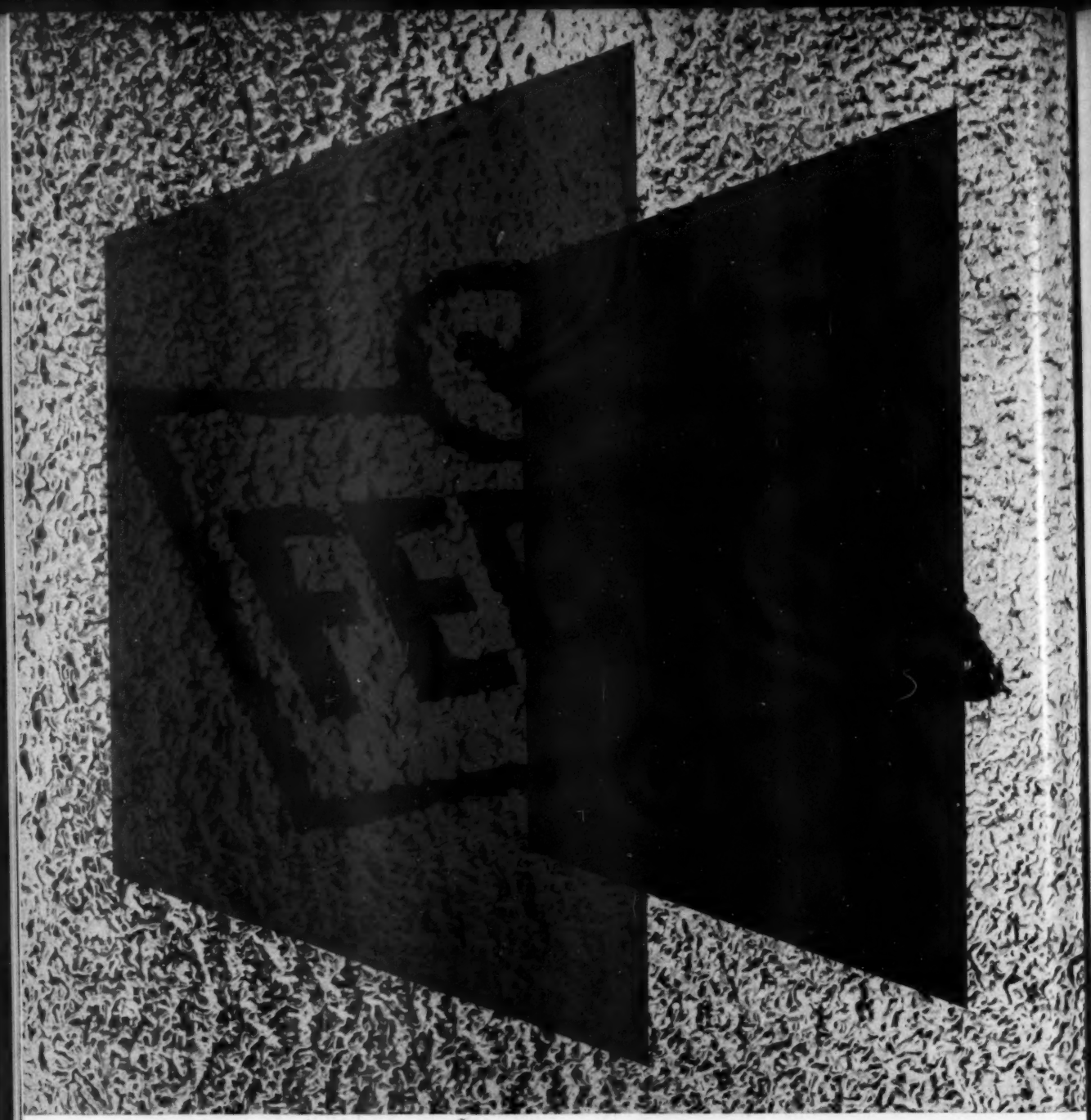
The importance is outlined of Raman spectra where the constitution of mol.-bonded compounds and methods for the experimental investigation are described. Results show that other than purely ionic bonds are existing in glass (ionic bonds are Raman-inactive). Characteristic vibrations of the groups SiO_4, BO_3, PO_4 and the effects exerted on them by the introduction of PbO and BaO in the glass structure are shown.

179. Ryosuke Yokota

"On the Structure of Soda-Silica Glass," J. Phys. Soc. Japan, 5, 354-56 (1950).

The "compound" theory is invalid as a means of explaining the atomic arrangement of soda-silica glasses. Yokota concludes from x-ray diffraction, specific volume, activation energy, and index data that silica glasses contain the configurations of the SiO_4 tetrahedra found in silicate crystals.

(Continued in December)



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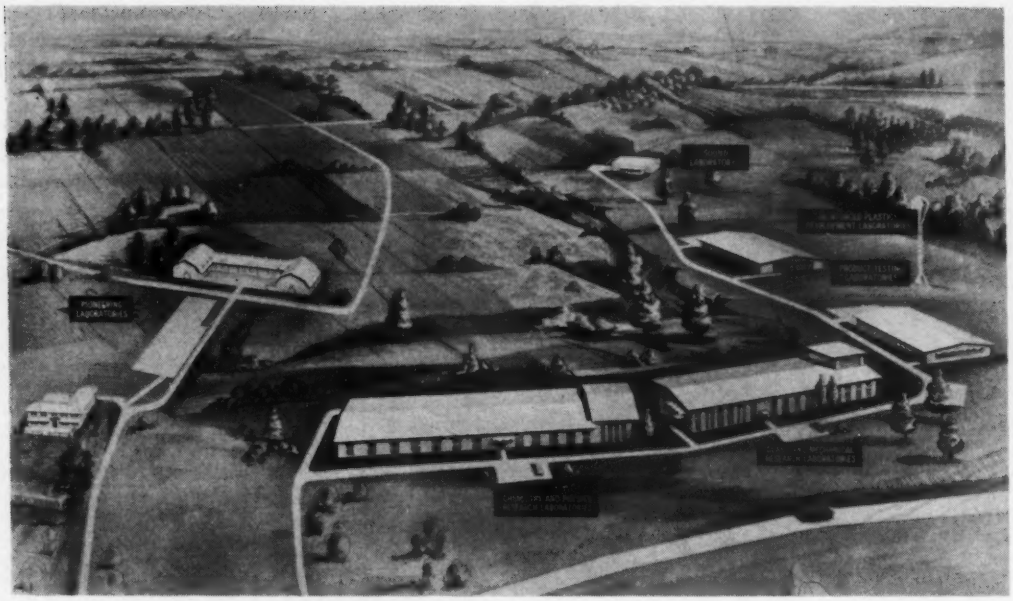


Owens-Corning Fiberglas Corporation's Technical Center

Owens-Corning Fiberglas Technical Center

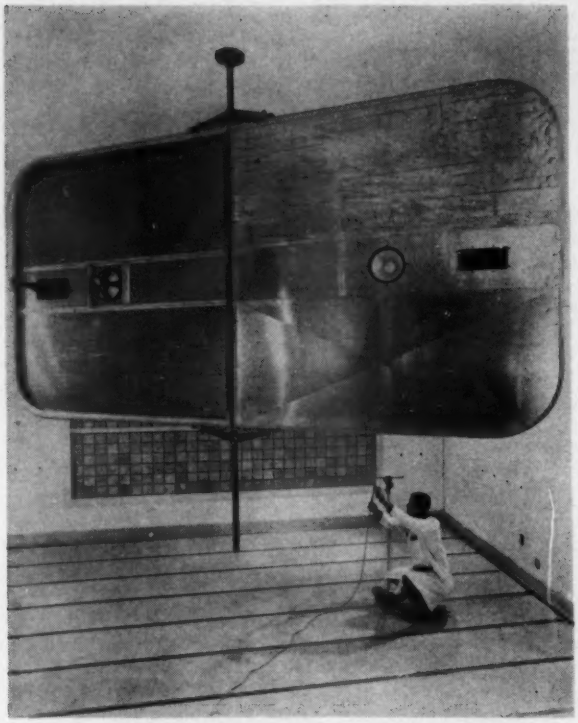
● DURING AN OPEN HOUSE INVITATION to members of the press last month, the complete and thorough organization of Owens-Corning's new Fiberglas Technical Center was at once evident. A multi-million dollar project, the seven research and testing buildings, have a total floor space of 154,000 square feet. The Center is a research composite in which more than 250 scientists, engineers, and technicians carry out laboratory studies on glass, reinforced plastics, product testing, and mechanical, metallurgical and acoustical research.

Research instruments include the spectrophotometer for the chemical analysis of solids, liquids and gasses; the viscometer for measuring the fluidity of molten glass at temperatures up to 2700°F.; the infrared spectrometer for determining the purity of new plastics; the metalloscope, for making photomicrographs of surfaces of high-temperature alloys to see if they will resist molten glass at fiber-forming temperatures; and the weatherometer, which subjects brightly colored fiber glass panels and other products to accelerated outdoor weathering tests.

The Pioneering Laboratory, a building by itself, is used for the advanced study of new products and processes. Considerable space and time are devoted to metallurgy since the successful production of glass and fiber glass depends on the use of metals at high temperatures.

Research work at the Center is being carried out under the guidance of Dr. August C. Siefert, Director of Research, and Dr. Games Slayter, Vice President in charge of Research.

An outstanding feature of the Research Center is the Sound Laboratory, very probably on a par with any other acoustical laboratory in the United States. The 12 inch thick concrete walls and two lead doors isolate the room mechanically from the rest of the building and a large vane, suspended from the ceiling, is positioned in



A 16-ft. vane in the Sound Laboratory. It breaks up sound waves to prevent standing wave pattern interference in acoustical tests. Sound absorption of acoustical materials and sound power output of noisy equipment are measured here.

its center. As sound is introduced into the room the vane rotates to provide a moving reflecting surface that prevents a fixed pattern of sound from developing.

The arrangement of the Sound Laboratory is such that three rooms are adjacent to one another and separated by walls having a fiber glass center core. The ceiling is made of acoustical ceiling board.

In the Chemistry and Physics Research Laboratories, scientists are seeking knowledge on the chemical activity of glass surfaces with the purpose of developing chemicals which will bond fibers together, thereby making glass more resistent to weathering and abrasion. It is here that the company produces new resins and chemicals on a pilot plant scale.

The Physics Laboratory is devoted to three general research areas. The first is concerned with the use of fibrous inorganic materials for use as a reinforcing medium in glass and other substances. The second is in fiber properties, and the third area is concerned with thermal properties and the flow of heat through insulating materials under varying conditions of temperature and pressure.

In the Glass and Mechanical Research Laboratories new glasses are under constant study and development. Studies are also carried on concerning new techniques for improving manufacturing processes concerning high temperature melting crystallization and other areas.

The staff of the Fiber Glass Reinforced Plastics Laboratory, in addition to developing new or improved reinforced plastics products offers technical assistance to fabricators and strives to develop new designs and processes of a practical nature. An interesting application of fiber glass throughout the Research Center is the appearance of fiber glass footings or bases under each of the machine tools. This permits the placement of precision tools near vibration tools with no danger of transmitting vibration from one to the other. The same holds true for an 85 ton air compressor.

In brief talks during a luncheon meeting, both August Siefert and Games Slayter stressed the great importance of fiber glass to markets in all fields. It was pointed out that fiber glass does not burn, is non corrosive, non-conductive and does not swell with moisture as there is no water absorption—an important factor in roofing materials. It is also possible to heat set fiber glass fabrics at high temperatures.

Commenting on the future of the fiber glass market, both Dr. Siefert and Dr. Slayter pointed out that there is no limit in sight and that the possibilities are even more exciting today than they were just a few years ago when the fiber glass market was in its first growth period.

The Research Center of Owens-Corning Fiberglas will be formally dedicated in May, 1961. However, it is now structurally complete and working at full staff.

Glass fibers formed at white-hot temperatures on an experimental device in the Pioneering Laboratory. Engineer checks for quality as molten glass is drawn through tiny holes in the glowing bushing to form the fibers.

NEW SLANTS ON HEAT PROCESSING FROM SELAS

Selas Gradiation® Heating minimizes annealing rejects at Wheaton Glass Company

Photo shows initial inspection as vials leave Selas-fired annealing lehr.

In a modern plant at Millville, N. J., The Wheaton Glass Company produces high quality tubing products, including ampules and vials. Each production line includes a 16-foot annealing lehr. Duradiant® burners in each lehr roof, only 18 inches above the conveyor belt, heat the products to the pre-selected annealing temperature.

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- freedom from contamination by incompletely burned fuels.
- conveyor belt life more than doubled!
- close temperature control within ± 5°F.
- no overheating and hence no deformed rejects.
- a very minimum of production time required for re-heating.

Wheaton Glass Company also has a 46-foot long lehr for fusing screened ceramic lettering on tubing-made containers. The fuel rate of each Duradiant burner is adjusted independently of the others so that the optimum heating pattern can be established through the lehr for the particular run. Here again, Gradiation heating provides correct fusing without concentrations of temperatures so high as to deform the carefully made glass containers.

If you would like to have specific details on Selas Gradiation heating and its applications in the production of glass and ceramics, write to Mr. C. P. Mann, Manager, Dryer, Oven and Lehr Division.

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CONFERENCE ON FIBER GLASS REINFORCED PLASTICS

PART II, Conclusion

High Temperature Plastic Materials for Missiles.

By C. S. Brown and Ray Cutler,

H. I. Thompson Fiber Glass Co.

For extremely high temperatures (5,000-15,000°F) to which missiles and some of their components are exposed, reinforced plastics perform better than most metals, a reverse of the situation for conventional aircraft. This reversal has come about because the times at temperatures are significantly less (2 seconds to 3 minutes), structural requirements are not as demanding, plastics offer excellent thermal insulation properties, and using the proper reinforcement and resin in the proper orientation minimizes the effect of ablation.

The mechanism by which Refrasil reinforced plastic resists degradation when used in nose cones and rocket engines exposed to high temperature, high velocities and high pressures is somewhat as follows.

The silica fiber has a melting point above 3000°F. Upon melting at the high temperature of exposure, the very high viscosity of the silica prevents it from being easily blown away. Also, the melted Refrasil vaporizes. This tends to cool the surface and contributes to a slow erosion rate. Since the plastic has a high insulation value, heat penetration is slow, and so is the melting rate.

Proper orientation of the fibers in the composite is very important. Considering the exit cone portion of a rocket engine, if the plies of the reinforced plastic in this area were laid one atop another like a parallel laminate, it would fail rapidly. The service conditions would carbonize the plastic at about 500°F and expose an entire ply of reinforcement, which would then blow off in large sheets or chunks. This action would be continuous, of course, with progressive delamination. However, if instead of parallel lamination an orientation were made, such as an overlapping shingle effect or with the fibers anywhere from 20 to 90° to the centerline of the nozzle, quite a different situation exists. Coming into the exit cone, the hot gases pass over ends of the fibers. Since the reinforced plastic material is a good insulator, only a slight amount of heat penetration occurs, which carbonizes the resin, and exposes the ends of the fibers. This exposure then allows the fiber ends to ablate away slowly and uniformly as the process is repeated over and over during the firing.

In an area of the rocket nozzle where great curvature and contour changes exist, the problem is solved by making the various areas as sections of optimum fiber orientation to be bonded together with high temperature adhesives, or by using chopped fibers with random orientation to prevent delamination and early failure by loss of chunks or sheets.

Development of a Glass Fiber Reinforced Firearm Barrel.

By J. W. Silva,

Winchester-Western Research Division, Olin Mathieson Chemical Corp.

A firearm barrel made of filament wound glass fibers impregnated with epoxy plastic over a thin steel tube has a density of 2.2, contains about 90 per cent by weight of glass, and has a hoop rupture stress well in excess of 200,000 psi.

In a gun barrel it gives a reduction of weight of 30 to 50 per cent compared with all-steel construction, and provides much greater protection against bursting when subjected to abnormal stresses such as result when the firearm is fired against an obstruction in the barrel. The epoxy finish is weather resistant and will not rust, fade or discolor. The barrel is less thermally conductive than is steel, is not cold to the touch in winter weather, and can be handled comfortably after fast round firing. The only steel in the gun is the steel liner, the fastening to attach the barrel to the gunstock, and the front sight.

Evaluation and Development of Glass Reinforced Plastic Pipe for the Petroleum Industry.

By R. M. Levy and B. M. Vanderbilt,

Esso Research & Engineering Co.

Laboratory and test programs indicate that high quality reinforced plastic pipe can be prepared from a number of resin types, such as epoxy, polyester and polybutadiene, and by a variety of techniques. Glass reinforced pipe appears to be the most promising of all new types available to replace steel pipe generally in the petroleum industry. Major deterrents to its use are believed to be (a) relatively high cost, and (b) inadequate pipe cou-

plings. When and if these problems are overcome, it is felt that the petroleum industry will find large volume uses for reinforced plastic pipe in its producing and pipeline operations.

Physical tests used include flexural, crushing, compression, creep, thermal expansion, pulsating pressure, vibration and sustained pressure.

Of the various tests carried out on the pipe itself, the sustained pressure test at three or more pressure levels is considered the most meaningful. Field tests using 2" diameter pipe up to a pressure of 1000 psi have indicated practical usage is feasible. A mechanical coupling, for use with large diameter pipe where threads are not practical, is under development. Use of a pipe liner of the thermoplastic or rubbery type is considered desirable when operating at pressures of 500 psi or more.

At present, Standard Oil Company (N.J.) affiliates have over 18,000 feet of glass reinforced pipe in service. Most is in low pressure gathering lines and salt water disposal systems. One line has shown to be far superior to steel in resisting paraffin deposition even while operating in –40°F temperatures. Another has been in salt water service at 900 psi for almost three years. In down hole applications some 1900 feet of line pipe with a well head pressure of 500 psi have been operating satisfactorily for a year and a half.

Reinforced Plastics in the Chemical Industry.

By J. J. Fisher and R. E. Gackenbach,

American Cyanamid Company

What was a hope ten years ago is now becoming a reality—reinforced plastics are becoming standard materials of construction for the chemical industry. This paper summarizes the results of five years of laboratory testing of glass reinforced chemical resistant polyester resins in various chemical solutions.

All of the tests were made on ⅛" thick laminates made of two ounce T-19 glass mat, room temperature cured in a press using a catalyst system of 2% peroxide paste and 0.1% diethyl aniline, followed by a postcure of two hours at 250°F. Effects were measured by determining flexural strength and weight changes after immersion, the specimens being weighed without drying and flexure tested in water at the exposure temperatures. A negative weight change indicates loss, and a positive one indicates absorption. Specimens were immersed in the various chemical solutions at 25°C for five years, and at 100°C or at reflux for up to one year.

The results are summarized in Table II.

At both room and elevated temperatures the greatest strength loss occurs in the first few months and the first few weeks, respectively, and then the strength-time curve levels off. Exceptions are exposures to nitric acid and NaOH, where strength continues to fall off with time after the initial drop in strength.

The laminates resist sulfuric acid up to 50% concentration, but not concentrated acid. They do not stand up to nitric acid. Even in 10% HNO_3 the strength continues to fall off after five years at room temperature.

Hydrochloric acid appears to be more corrosive than phosphoric acid but there is adequate strength retained in the former to permit its use in the hydrochloric acid.

The resistance to 10 and 30% sodium hydroxide at 25°C is less severe than might be expected, but the NaOH becomes very corrosive as the temperature increases. Not only is there a great loss of strength, but the weight change is dramatic. Aluminum sulfate (alum) causes little weight change under any conditions, and less strength loss than either inorganic acids or alkalis.

Organic acids and formaldehyde, with the exception of glacial acetic acid, should not cause any problems with a chemically resistant polyester binder. In organic solvents there is a noticeable absorption, but loss of strength is limited. Only benzene continues to cause strength loss after five years.

The general conclusions are: (1) oxidizing acids are

Table II

EFFECTS OF LONG TERM EXPOSURE OF FIBER GLASS-POLYESTER LAMINATES IN VARIOUS CHEMICALS

	Weight Change (%)		Flexural Strength, psi	
Chemicals	Five Years at 25°C	One Year at 100°C or as noted	Five Years at 25°C	One year at 100°C or as noted
10% H_2SO_4	3.3	2.31 (2 mo.)	12,500	3,000 (7 mo.)
50% H_2SO_4	1.7	1.61 (7 mo.)	13,000	1,500 (7 mo.)
10% HNO_3	—12.0	— 4.9 (1 mo.)	4,800	4,000 (1 mo.)
Conc. HNO_3	28.2 (1 yr.)	.....	1,500 (6 mo.)	.....
10% HCl	—10.4	—12.0 (4 mo.)	4,200	2,800
20% HCl	—10.4	—14.0 (4 mo.)	3,600	2,400
10% H_3PO_4	— 6.3	— 2.5	8,000	3,800
Conc. H_3PO_4	1.1	— 1.8	10,500	5,000
10% Alum	0.3	0.1	16,000	7,000
Satd Alum	0.4	0.4	17,000	6,000
10% NaOH	— 0.6	13.6 (2 wk.)	4,800	4,500 (2 wk.)
30% NaOH	— 3.5	8.5 (1 wk.)	4,800	4,000 (1 wk.)
40% Maleic acid	0.9	— 3.5	12,500	4,700
10% Acetic acid	1.2 (1 yr.)	— 3.6	13,500	6,900
Glacial " "	13.6	1.2	13,000	4,200
37% Formaldehyde	3.8	— 2.9	18,000	6,500
Acetone	9.5	.....	12,500	.....
Methanol	5.9	.....	14,000	.....
Benzene	12.9	.....	10,500	.....
Water	0.34	— 2.1	14,000	4,200

more corrosive than organic acids, water, and inorganic salts, (2) organic solvents attack more through absorption than decrease in strength, and (3) a chemically resistant polyester laminate can lose from 25 to 75% of its original dry strength over a five years period of immersion at room temperature.

Typical polyester chemical equipment in use includes vats, tanks, ductwork, hoods, piping, trays, filter plates, towers, fume scrubbers, fans, and blowers, as well as tank lining and floor coverings.

The Deterioration of Epoxy Laminates Under Extreme Aging Conditions.

By N. C. W. Judd, T. Lloyd, P. McMullen and E. W. Russell

Royal Aircraft Establishment, Farnborough, England

This study was made to determine the cause of the severe loss in strength of certain reinforced plastic structures, made of Volan treated glass rovings and epoxide resin, which had been stored several months at 100% relative humidity and 50°C. The failure of tensile test pieces was of the brittle type, differing from that observed with polyester systems or unaged laminates, and suggesting some type of attack on the glass reinforcement.

The extensive study disclosed that the deteriorating effect appears to depend on a number of interdependent factors, chiefly those relating to the surface condition of the glass fibers. The strength loss can be reduced by the use of acid anhydride curing agents of certain silane surface treated finishes on the glass, and some combinations of these materials give laminates with a high resistance to the deteriorating effects of hot moist storage.

Glass Reinforced Plastic Laminates in Contact with Sea Water or Fuels.

By J. B. Alfers and W. R. Graner,

Bureau of Ships, Department of the Navy

A variety of tests have been conducted by the Navy to determine the effects of different service environments upon glass reinforced plastic over a period of years. Five types of glass reinforcement in polyester laminates exposed to sea water immersion at Kure Beach, N.C. for four years showed very good strength retention properties. The best samples were those of Garan finished glass cloth or mat which retained over 90 per cent of original flexure strength.

This ability of glass reinforced polyester laminates to withstand exposure to sea water is confirmed by tests made on the fairwater of the U.S.S. Halfbeak after five years in service. The material retained 88 per cent of its flexural strength and 91 per cent of its flexural modulus in actual submarine service which included high wave slap loadings, repeated exposure to high submergence pressures, and repeated wetting and drying cycles in sea water. Of interest, the specific gravity of the material increased from 1.68 to 2.66 over the five-year period, but the paper gives no analysis of the change.

Exposure tests of a series of nine polyester and five epoxy resin laminates in aviation gasoline, jet engine fuel, marine diesel fuel and Navy Special fuel oil for six months disclosed that, aside from aviation gas, the fuels were not affected by the laminates (as measured by gum tests). Those samples that did not pass the gum test in aviation gas were rerun with samples made by better techniques and the latter passed the tests.

The fuels affected some samples, especially the fuel oil. However, samples made of low exotherm polyester and epoxy resins satisfactorily resisted all four fuels.

The Design of Reinforced Plastic Compositions for Impact Resistance.

By E. G. Bobalek and R. M. Evans

Case Institute of Technology

This report deals with part of an extensive program directed to search for the chemical variables of composition and structure which upgrade the impact resistance of a variety of plastics, both thermosetting and thermoplastic. The part presented is work dealing with the interaction between fibrous reinforcing fillers and thermosetting resin.

In general gains in impact resistance can be achieved most easily by increasing elongation at the expense of some rigidity. Both the tensile strength and modulus of laminates made of resin reinforced with continuous fabric can be roughly approximated as the algebraic sum of the strengths and moduli of the volume fractions of the two constituents. Since the strength of both glass and nylon fabric is roughly 15 to 40 times greater than that of any non-reinforced resin, increasing the volume fraction of the fibrous filler rather than reformulation of the binder is the most practical way to increase strength.

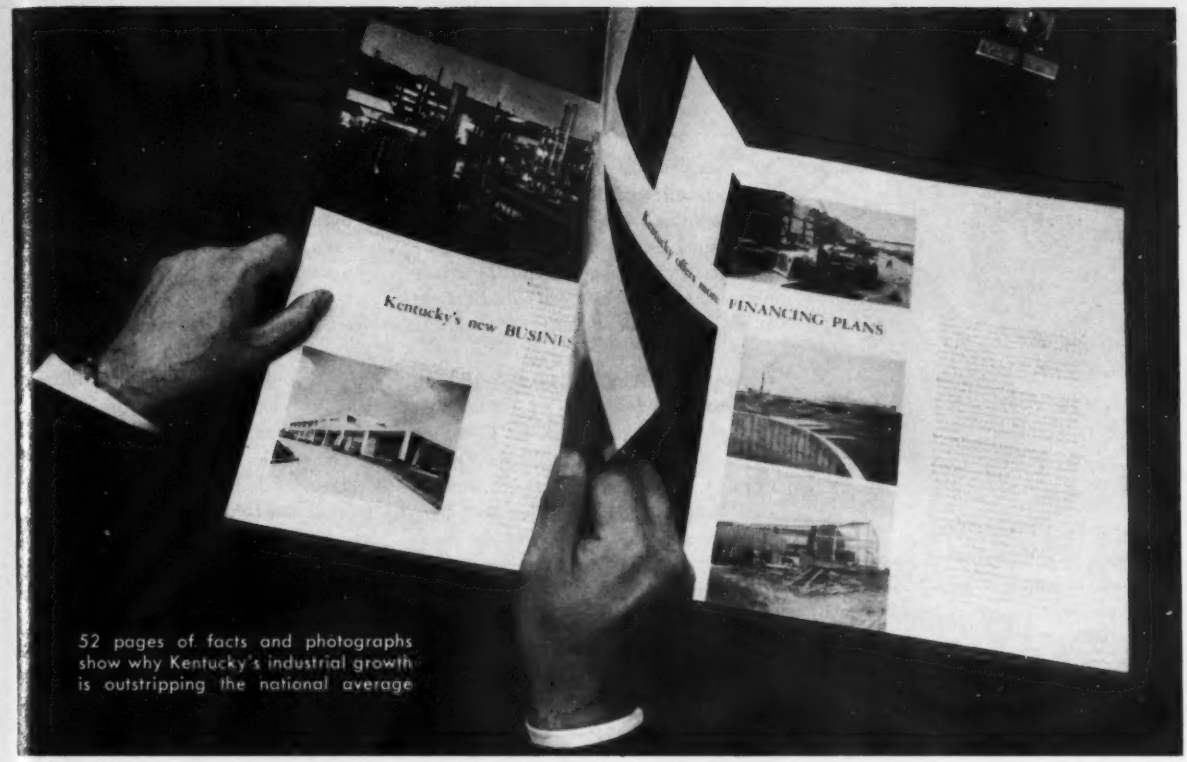
Advantages of Producing a Truck Cab from Molded Fiber Glass.

By R. L. Wiese

Molded Fiber Glass Body Co.

Fiber glass reinforced plastic (molded fiber glass) has seven advantages for the production of truck cabs and automobile bodies:

1. Low tooling cost—$39,000 against $240,000 for sheet steel in one type of truck cab.
2. No rust or corrosion—a Chevrolet Corvette body has been driven 125,000 miles since 1954 with no rust or corrosion
3. High impact strength—a molded part can be deflected to a point just short of its yield strength and return to its orginal shape without damage and with no permanent deformation.
4. Ease of repair—impact damage is confined to the point of contact, and the damaged area can easily be replaced in the truck company garage.
5. A quiet assembly—joints which are 100 per cent bonded do not rub, squeak or rattle and sound transmision is low.
6. Reduction in weight—0.100″ thick molded fiber glass weighs 0.90 pounds per square foot as compared with 1.5 for 20 gauge steel (0.035″ thick), both thicknesses being those used in truck cabs.
7. Unlimited design possibilities.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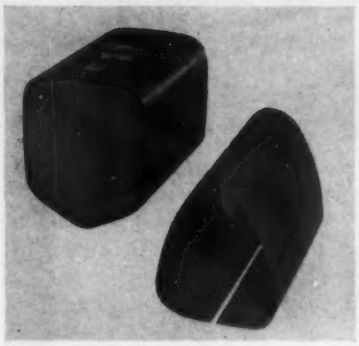


Contoured Fiber Glass

The permanent formation of flat molded fiber glass panels into extreme and compound curvatures, without expensive molds or a sacrifice of physical properties, is being accomplished by the Molded Fiber Glass Sheet Company, Ashtabula, O. Instrument and machine housings, containers and tanks are the main products of the new process at the present time. Curvatures down to a radius of 3 inches and any wall thickness are possible.

Fiber Glass Report

The September Monthly Letter of Goodbody & Company contains a detailed study of the fiber glass industry that found fiber glass to be one of the nation's fastest growing raw materials. It states that consumption of textile glass fibers, which since 1950 has grown at a rate of 22 per cent compounded annually, rose 37 per cent in the first half of 1960 over the first half of 1959. An estimated capacity of 318 million pounds is expected for textile fibers in 1961 and industry sources estimated the 1970 demand to be 700 million pounds. Appeal for long-term investment is still evident despite a substantial rise in fiber glass companies' stocks.

Gustin-Bacon Earnings Rise

Earnings at Gustin-Bacon Manufacturing Company for the fiscal year ended September 30, rose to over $2,300,000, or about $1.60 per share, on sales of approximately $32 million compared to the 1959 fiscal earnings of $2,082,386, or $1.43 per share, on sales of $28,655,908 million.

Output from a $6 million expansion program is expected to start at the beginning of the year. The primary products affected by the program are fiber mat and board products for the residential home, auto and appliance markets. Production capacity in textile fibers for the reinforced plastic field is also being increased.

Owens-Corning Fiberglas Ups Expenditures

Capital expenditures will be increased by 10 per cent to $44 million at Owens Corning Fiberglas. The announcement was made by Harold Boeschenstein, president, at the opening of the company's new technical research center, Granville, O. (See page 635)

Designers Commissioned

Pittsburgh Plate Glass Company's Fiber Glass Division commissioned five of the world's leading fashion designers to provide luxury patterns with entirely new design concepts for fiber glass drapery materials. The designers are Tom Brigance, Hannah Troy, Ceil Chapman, Pauline Trigere and Adele Simpson. The plans also include five major drapery firms, Charles Bloom, Inc., Qual Fab Inc., Covington Fabrics, Titus Blatter, and Waverly Fabrics.

New Distributor

The second quarter of next year will see Iselin-Jefferson Co., Inc., and Shelby Mills, Inc., producing and distributing fiber glass decorative fabrics in the novelty and high style line. Manufacturing space for a modern coronizing unit to process fiber glass yarn is now ready at Shelby Mills. Additions and alterations are in progress on warping, slashing and weaving equipment to adapt them to the manufacture of glass fiber fabrics.

Union Contract

A new three year contract between Johns-Manville Fiber Glass Inc., and the Glass Bottle Blowers Association of the United States and Canada, A.F. of L.-C.I.O, Local 54, provides a substantial wage increase to all newly evaluated rates, effective as of November 1, 1960. The Union represents the hourly employees at J-M's Waterville, O., plant and technical center.

In addition to wage increases a new retirement program was adopted, improved vacation and insurance benefits were granted and a new job evaluation program was agreed upon.

Hi-Strength Fiber Glass

Fiber glass with 50 per cent greater rigidity and no sacrifice in strength has been developed for the Air Force by Owens-Corning Fiberglas.

The fibers' prime application is where increased stiffness is required in reinforced plastics. Named "High Modulus Glass," it can be used in the development of aerospace vehicles and missiles to permit important weight savings. A potential use for immediate application is in reinforced plastic filament wound rocket motor cases.

Over 62 different glass compositions were investigated at Owens-Corning's Technical Center, Granville, O., before one was found that would meet the Air Force requirements established by the Wright Air Development Division, Air Research and Development Command, Dayton, O. The announcement of the new glass fiber was made at a technical meeting in Dayton, October 12, where six papers on the subject were presented. The papers, listed below, are available from Mr. George P. Peterson, WWRCNC-2, WADD, Wright-Patterson Air Force Base, Dayton, Ohio.

1. *Properties of High Modulus Reinforced Plastics.* George P. Peterson, Wright Air Development Division.

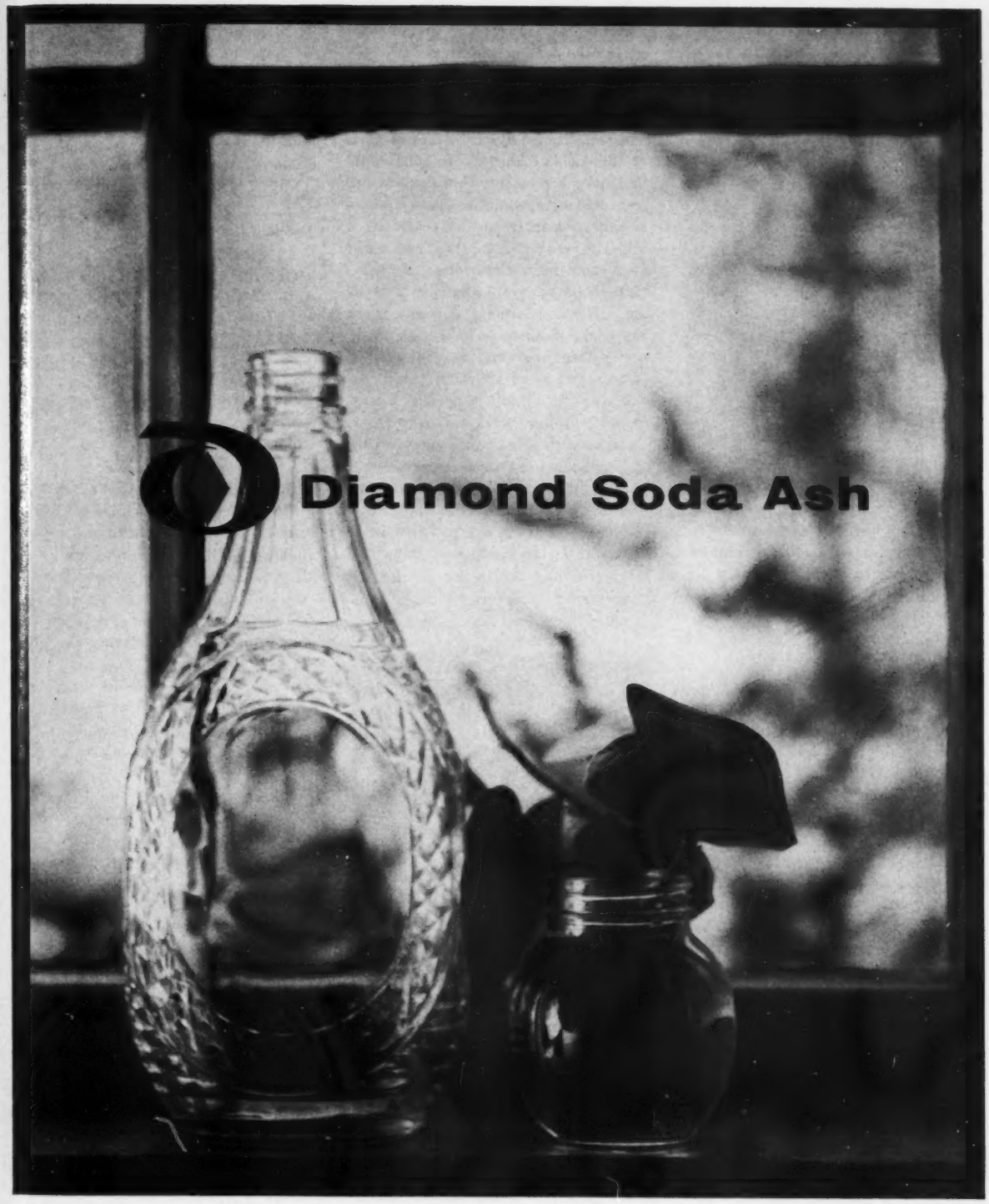
2. *High Modulus Glass Fibers for Structural Plastics.* R. Tiede, Owens-Corning Fiberglas, Corp.

3. *Processing and Fabrication Techniques for Reinforcing Plastics for YM 31 A Fibrous Glass.* A. Isham, Owens-Corning Fiberglas Corp.

4. *Structural Analysis.* E. Kuenzi, Forest Products Laboratory.

5. *Safety Precautions for Handling and Fabrication.* R.L.H. Miller, Kettering Laboratory.

6. *Potential Uses of High Modulus Glass Fibers.* P.L. Layton, Owens-Corning Fiberglass Corp.



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DIAMOND ALKALI COMPANY, Union Commerce Building, Cleveland 14, Ohio

Diamond Chemicals®

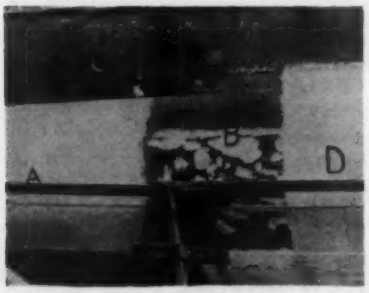
APPLICATIONS

(Fig. A)

FIBER GLASS FLAKE has been applied to the corroded and pitted steel hull of the *Joseph Conrad,* a square rigged sailing ship (Fig. A) used for a youth-training program by the Marine Historical Association at Mystic Seaport, Conn. A typical New England seafaring community, complete with vessels, has been restored by the Association at Mystic.

The *Joseph Conrad's* hull, particularly below the water line, was in such a state of deterioration that in some areas the welding of patch plates was impractical. Estimated cost per new plate exceeded $1,000. Therefore the idea of encasing the entire hull in fiber glass at a considerable savings over the cost of even partial replating was considered more feasible.

A 200 sq. ft. test patch of fiber glass flake was applied last winter and the ship was left in tidewater from October until May, then drydocked and inspected. The protective coating was found to be securely in place and behind it the hull's iron plates were "bone dry."



(Fig. B)

Area "A" (Fig. B.) is the fiber glass flake coating and areas "B", "C", and "D", represent other fiber glass coatings.

The new coating covers the ship's 4,600 sq. ft. of hull with flakes of glass of pin-head diameter. Infinitesimally thin, they are combined with polyester resin and sprayed layer upon layer in a pattern similar to the scales of a fish. The result is ⅛-in. thick coating of glass and resin that forms a tough, virtually impenetrable barrier to moisture. The overlapping laminations of glass flake produce an inert, chemically stable "multi-barrier" to the passage of corrosive gases which are more destructive and difficult to hold back than corrosive solids, such as sea water. Tests have revealed vapor transmission of less than .01 perms, shear strength of 800 psi. and dielectric strength of 500 volts per mil. The same application of fiber glass flake was put on one of Alcoa Steamship Company's ships last May. It is engaged in the bauxite ore trade between Dutch Guiana and Trinidad.

(Fig. C)

In the actual application a positive pressure spray system that moves solids from pot to nozzle without atomization is used. The fiber glass flake, factory compounded with the polyester resin, and the activating agent are drawn from separate, sealed pots by the spray system, mixed in the proper proportion, and sprayed from the nozzle of a gun to the work. This distance from nozzle to work is usually about 18-inches which minimizes the inducement of air. (Fig. C)

The two man crew can spray 300 to 400 sq. ft. an hour and colors can be easily added to the resin-glass mix for decoration and to insure 100 per cent coverage on the second coat. The second coat follows after the first has cured, about one hour.

Before the application, the surface is thoroughly cleaned by sand-blasting or wire brushing to insure a clean, stable surface.

The *Joseph Conrad's* hull is black, the bulkheads are white, and water line to water line is red.

Fiber Glass: Owens-Corning Fiberglas Corp., 717 Fifth Ave., New York 22, N.Y.

Spraying Equipment: Glass Plastics Corp., Linden, N.J.

MOLDED FIBROUS GLASS is used to thermally insulate lighting fixtures for incandescent bulbs. Designed to serve with parabolic reflecting surfaces such as are conventional in this type of installation.

Fabricator: Fibrous Glass Products, Alpa Plaza, Hicksville, L.I., N.Y., subsidiary of Pall Corp.

FIBER GLASS ABRASIVE BLADE for use on concrete blocks of all types; refractories, including medium to high alumina, mullite, sillmanite, basics of all types and some super duty brick and building materials such as sandstone, pre-cast units and certain asbestos-cement products. To protect the fiber glass cloth from becoming thin with side wear silicon carbide particles saturate the cloth and protrude beyond its surface.

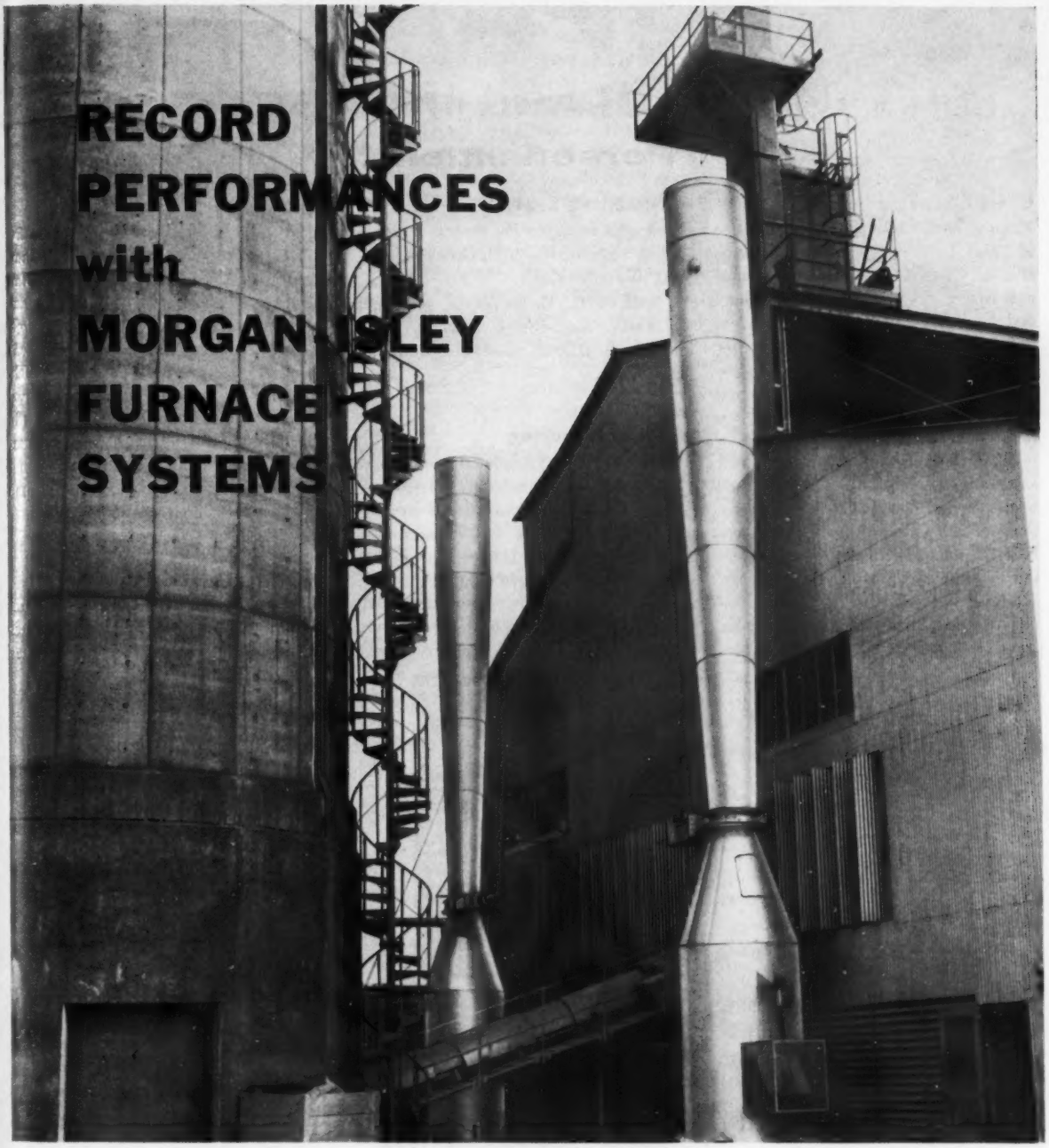
Fabricator: The Clipper Mfg., Co., 2800 Warick, Kansas City 8, Mo.

FIBER GLASS DOME cover combats corrosion in the 25 ft. diameter broke chest of a southern pulp and paper mill. Helically wound stainless steel cables imbedded around the circumference provide extra reinforcement to hold the cover in compression when loads are exerted. The cover was molded in eight pie-shaped segments to facilitate shipment then assembled at the paper mill.

Fabricator: Metal-Cladding, Inc., North Tonawanda, N.Y.

FIBER GLASS INSTRUMENT CASE COVER is being used on a pneumatic transmitting bellows flow meter. The weatherproof case is 9″ high by 7″ wide and 4″ deep. It is die-cast aluminum with a reinforced fiber glass cover. The pressure meter is used to measure liquid level in closed tanks under pressure or vacuum, and for flow or open tank measurements.

Fiber Glass: Owens Corning Fiberglas Corp., Corning, N.Y. Instrument: Brown Instrument Div., Minneapolis-Honeywell Regulator Co., Wayne and Windrim Aves., Philadelphia 44, Pa.



Illustrated above is a MORGAN-ISLEY SYSTEM with secondary regenerators at United Can and Glass Company, Hayward, California. This furnace melted a record-breaking 254,036.6 tons in 1773 operating days with a melting area of 676 square feet and an average fuel consumption of 5979 cubic feet (natural gas) per ton. The Morgan-Isley system can help you approach record-breaking performances.

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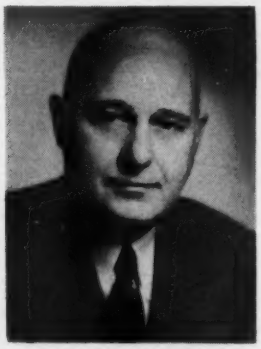
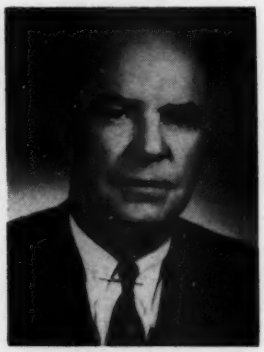
NEWS
IN THE GLASS INDUSTRY
Personalities...

C. G. Bensinger
Member of the Management Committee of nine operating division, Owens Illinois Glass Company. Bensinger is executive vice president of the company and he will continue as a member of the Corporate Planning Committee. He has been president of the Glass Container Division since 1956 and served the Division in other executive capacities since 1931.

R. H. Mulford, vice president, will assume the responsibilities relinquished by Bensinger. They include the operations of the Glass Container Division-East, the Pacific Coast Division, and the Closure and Plastics Division. Mr. Mulford headed Kimble Glass Company, a subsidiary of Owens-Illinois, from 1953 until last month.


C. G. Bensinger


R. H. Mulford

Thomas M. Huber

Alden P. Taber
Vice president and head of the research and development division, Babcock & Wilcox Company.

Maurice L. Stonehill
Elected president, Jeannette Glass Company following the resignation of Burt K. Todd.

Keith B. Fackler
And Carl H. Seyfer, field representatives, have been appointed to the distributor sales organization, Chicago district, Libbey-Owens-Ford Glass Company. Frank C. LaVrar and Robert B. Sarver have been assigned to the Atlanta and Minneapolis district offices, respectively.

William C. Lewis Jr.
Research manager for process development, Ceramic Research Division, Corning Glass Works.

Wilfred L. McMahon is personnel manager, Engineering Division. . . . Ernest E. Russell, manager, Refractories Plant.

William Manring
Joined the Chlor-Alkali Division, Food Machinery and Chemical Corporation as head of the technical service department for the glass industry. He was formerly vice president of Glass Technicians, Inc., Hamilton, O. Prior to that position he served as chief chemist for Ball Brothers Glass Company.

John W. Thayer
Marketing manager, Metro Glass, a division of National Dairy Products Corporation.

James H. Joyner
Manager, Hawaiian sales, H. K. Porter Company, Inc. The product lines include refractories.

J. James Offutt
Vice president and general manager of A. P. Green Fire Brick Company. He will be responsible for the direction of all manufacturing, sales, research and engineering.

M. D. McClain has been appointed assistant to the general manager. . . . Jack W. Swate, director of sales. . . . H. L. Beynon, general sales manager.

Francis W. Theis
Elected president and chief executive officer of Pittsburgh Plate Glass Company International, S.A. Bjorn Holmstrom is now chairman of the board of directors. Mr. Theis succeeds Dwight R. Means who is retiring but who will continue to serve as a consultant. The company, a subsidiary of Pittsburgh Plate Glass Company, is engaged in the sale of flat glass, fiber glass, paint and chemical products, and in investing, licensing and manufacturing operations, outside the United States.

Thomas M. Huber
Vice president, Owen-Illinois International S. A., with responsibility for administering technical assistance and licensing agreements overseas. Mr. Huber will also investigate new international investment opportunities in areas outside of Europe, the Middle East, and South America.

He was formerly manager of the Owens-Illinois glass container plant in Cuba until the recent intervention of the Cuban Government.

Clayton F. Devine
Executive vice president and member of the board, Ottawa Silica Company.

Eugene A. Fischer
Sales manager of refractories, Norton International, Inc. Gale W. Bennett has been named assistant manager of product engineering, Refractories Division.

Richard M. Smith

James M. Ashley

has been elected director emeritus of the Producers' Council, an organization of building material manufacturers, in recognition of his service to the council over the years. He is vice president of public relations, Libbey-Owens-Ford Glass Company.

Since the Producers' Council was founded in 1921 there have been only two other directors emeritus: Tyler S. Rogers, formerly of Owens-Corning Fiberglas, and L. S. Hart, Johns-Manville.

A. M. YOUNG, general sales promotion manager, was elected a director of the council.

Richard M. Smith

Plant manager for Corhart Refractories Company's new refractory plant, under construction at Buckhannon, W. Va. Corhart is a wholly-owned subsidiary of Corning Glass Works.

Smith joined Corning Glass Works in 1950 as junior process engineer at the company's refractories plant. Since then he has held the positions of senior process engineer, supervisor of quality control and production planning, production superintendent, and, since 1958, plant manager.

The new operation at Buckhannon is expected to begin manufacturing next spring. (See drawing below.)

Paul B. Stewart

Vice president, Owens-Illinois International. He will be succeeded as general manager of O-T International's Venezuelan glass container plant by Dexter W. Childs, who has served as the plant's administrative manager.

The Venezuelan plant is completing an expansion program which will more than double its initial capacity. Plastic closures will soon be added to its production line which now consists of glass continers for Venezuela's food, drug, beverage and chemical industries.

Peter Colefax

Peter Colefax

Chairman of the board, American Potash & Chemical Corporation. He will also continue as president of the company.

T. J. Pohl

Sales manager, Insulation Division, M. H. Detrick Company.

Karl R. Wuensch

Named plant manager of new electronic component plant which is to be built in Raleigh, N.C., by Corning Glass Works. He has served as production superintendent at Raleigh for three years.

Companies...

Kaiser Refractories & Chemicals Div.

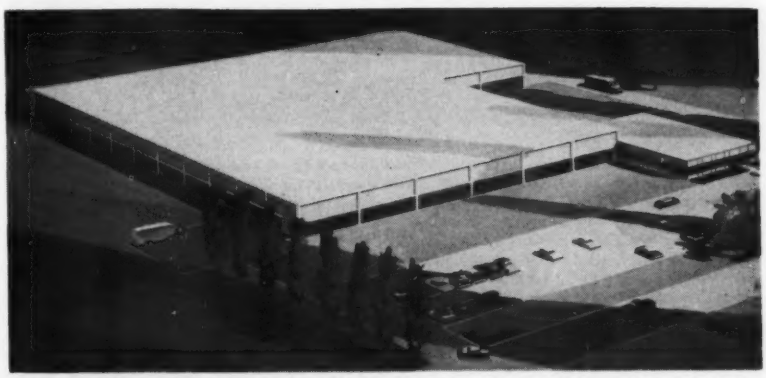
Constructing a new Technical Center at Mexico, Mo. The Center will be equipped to test and evaluate material of all types in connection with the development of fireclay, silica and high alumina refractories in the form of brick, mortars, castables, plastics or ramming mixes. The building will also be the headquarters for the Division's raw materials department.

Electronic Expansion

A new plant for the manufacture of electronic components will be built at Raleigh, N.C., by Corning Glass Works. The 120,000 sq. ft. structure will be situated on a 32-acre site and will be devoted initially to electrical capacitors for use in electronic circuits. Capacitors are presently manufactured at the company's Bradford, Pa., plant but this plant will concentrate on other electronic components after the new facility begins operating next summer.

Corning has also announced plans for a 400-acre research and engineering center near Corning, N.Y. A pilot plant will be the first of a series of buildings to be erected during the next five years. It will house nine melting furnaces and two process research departments with their special tools and equipment.

The group of buildings will be called the Eugene C. Sullivan Science Park in honor of Dr. Eugene C. Sullivan, honorary chairman of the board, and founder of the company's research laboratories over 52 years ago. The company allots about four per cent of its sales dollar to research. Last year's expenditure reached a record high of $8 million.



Artist's drawing of Corhart Refractories Company's new refractories plant, Buckhannon, W. Va.

Feldspar Corp.

Will be in a position to ship feldspar from its newly constructed Middletown, Conn., plant before the end of this month. The operation will have a capacity of 7,000 tons of feldspar per month and will also produce silica, silica flour and mica as by-products.

New Mirror Freight Classification

Members of The National Furniture Conference received the following bulletin from their organization:

"Supplement 12 to Uniform Freight Classification 5, effective September 1, 1960, includes a new 'F' package — PACKAGE 93-F — applicable on mirrors, under Item 42356-A, Note 8, of UFC 5, and Item 42570-A of UFC 5.

The specifications of this package read as follows:

1. Mirrors must be individually packed in containers not exceeding 85 united inches, gross weight not exceeding 50 pounds, made of double-faced corrugated fibreboard testing not less than 200 pounds.

Or in containers not exceeding 100 united inches, gross weight not exceeding 70 pounds made of double-faced currugated fibreboard testing not less than 275 pounds.

Or in containers not exceeding 100 united inches, gross weight not exceeding 75 pounds made of double-faced corrugated fibreboard testing not less than 350 pounds. Containers must have full overlap construction top and bottom.

2. Mirrors must be protected with blankets or pads as described in Item 42352, Note 4, Paragraph (1), folded and arranged to maintain not less than 3/4 inch clearance between all finished surfaces of article and inside walls of container. Folded blankets must incase the ends of the mirror from top to bottom and not less than one folded blanket or pad must span the face of the mirror in the center from top to bottom and in no case shall folded blankets or pads be more than 12 inches apart. Blankets or pads must be securely held in place.

3. Containers must be closed in compliance with Rule 41, Paragraph 7."

Mirror Manufacturers Assn.

The National Association of Mirror Manufacturers has established its headquarters in Washington, D. C., and James E. Mack has been appointed Administrator. This Association has no connection with the now dissolved Mirror Manufacturers Association.

The new Association's principal objective will be to advance, promote and protect the mirror manufacturing industry. One of its first activities will be to oppose a proposal to further reduce the import duty on mirrors. It will present the industry's case in opposition to such a reduction at a Tarriff Commission hearing, next year.

The organization's annual meeting was held on October 10-11 at the Mayflower Hotel, Washington, D.C. The meeting included an address by Federal Trade Commissioner Sigurd Anderson regarding lawful and unlawful trade practices under the Robinson-Patman act.

Members of National Association of Mirror Manufacturers at annual meeting on industry problems, October 10 and 11 in Washington, D.C. Clockwise, starting at end of table in foreground are: Edd Gardner, Carolina Mirror Corp., N. Wilkesboro, N.C.; Jack Helms, Weaver Mirror Co., Rocky Mount, Va.; J. A. Morse, Toledo Plate & Window Glass Co., Toledo, O.; W. B. Walter, Toledo Plate & Window Glass Co., Toledo, O.; A. G. Jonas, Jr., Lenoir Mirror Co., Lenoir, N.C.; John Messer, Jr., Galax Mirror Co., Galax, Va.; Paul Bertell, Standard Mirror Co., Buffalo, N.Y.; Arthur Bienenfeld, Century Industries, Chicago, Ill.; Milton Rubin, Metropolitan Mirror & Glass Co., Mount Vernon, N.Y., Association executive secretary James E. Mack; and Association president Kay Willard, Willard Mirror Co., Fort Smith, Ark.; Association secretary-treasurer R. H. Turner, Falconer Plate Glass Corp., Falconer, N.Y.; William Beeler, Virginia Mirror Co., Martinsville, Va.; Hy Tyre, Tyre Brothers Glass Co., Los Angeles, Calif.; Charles Younce, Lenoir Mirror Co., Lenoir, N.C.; Ben Newton, Semon Bache & Co., New York, N.Y.; Marshall Lewis, Binswanger Mirror Co., Memphis, Tenn.; John Fritz, T. C. Esser Co., Milwaukee, Wis.; Larry Esser, T. C. Esser Co., Milwaukee, Wis.

Indian Sheet Glass Expansion

According to present indications there will be considerable expansion in the Indian sheet glass manufacturing industry, during the Third Five Year Plan period.

Presently there are four sheet glass manufacturers in the country with a combined annual production capacity of 106,560,000 square feet. Production in 1959 was 80,565,600 square feet.

The capacity for production for all types of glass and glassware, including opthalmic glass, as of March 31, 1961, will be 457,000 long tons, according to the draft outline of the Third Five Year Plan released by the Government of India. This capacity is expected to be increased to 615,000 tons by the end of the Third Five Year Plan period. The anticipated increase in the demand for glass and the favorable attitude of the Government of India toward expansion in glass production may provide worthwhile investment opportunities to interest American manufacturers for collaboration with Indian industrialists and businessmen.

British, Belgian, and Japanese firms are already associated with the Indian glass industry according to available information. Sheet glass is assessed an import duty of 70 per cent ad-valorem, in accordance with the recommendations of the Indian Tariff Commission which investigated the problems of this industry in 1950, 1954, and 1957. The industry has requested for a continuation of the present duty beyond December 31, 1960. The request is now under consideration by the Commission. Imports of sheet glass including plate glass are restricted considerably and the import quota fixed for the licensing period April to September 1960, for established importers, was 10 per cent.

According to an industry source in India, the Ministry of Commerce and Industry of the Government of India has issued licenses to six other firms for manufacturing various types of sheet glass including wired and figured glass which are not currently produced.

The demand for special types of glass such as safety wired and figured glass, for use in railroad coaches, windshields, and other purposes, is steadily increasing. The domestic demand for fiber glass is also expected to be high in the coming years—*Consumer Durable Goods Division, Business and Defense Service Administration, U.S. Department of Commerce.*

ACS-Pittsburgh Section

The annual joint meeting of the Pittsburgh Section of the American Ceramic Society and the Pennsylvania Ceramics Association was held Tuesday, October 11.

The program on the subject "Guidance Counselors — This is Ceramics," was divided between Harbison-Walker's Garber Research Center tour and industrial exhibits in the afternoon, and speakers at the Mellon Institute in the evening.

New board members for the Pennsylvania Ceramics Association are: Richard A. Jacobs, vice president and plant manager, Brockway Glass Company; Roland Van Der Beck, plant manager, Carborundum Company, Latrobe; Horace L. Latimer, The O. Hommel Company; and Joseph A. Reising, St. Joseph Lead Company.

Glass Symposium in Italy

A Symposium on the Mechanical Properties of Glass and Methods of Improvement will be held in Florence, Italy, in September, 1961, according to P. Migeotte, secretary of the Union Scientifique Continentale du Verre. Arrangements for presenting papers should be made without delay. Additional information is available from the office of the Secretariat, Union Scientifique Continentale du Verre, 24, rue Dourlet, Charleroi, Belgium.

Texas Gulf Sulphur Co.

Plans to invest $20,000,000 to $30,000,000 in the development of rich potash reserves in Utah. Completed core drilling has found the reserves to be richer than any others known to exist in the United States.

The president of Texas Gulf Sulphur, Claude O. Stephens, said he would "strongly recommend" that his directors authorize a diversification into potash, "if studies nearing completion yield final reports as favorable as all interim reports have been."

The company indicated it might build a plant with a 10,000 ton capacity per day for processing potash.

An option to buy the reserves near Moab, Utah, from Delhi-Taylor Oil Corporation would give Delhi-Taylor a 25 per cent interest in net profits from the operation. Delhi-Taylor will also receive guaranteed advance profits of $4,500,000 over a four and one half-year period. Texas Gulf Sulphur has until January 1 to complete its evaluation of the deposit. It has already spent more than $500,000 on its evaluation of deposits at Cane Creek, in southeastern Utah.

Listed for Trading

The common stock of Engelhard Industries, Inc., refiner and fabricator of platinum metals, was listed for trading last month on the New York Stock Exchange. It opened at 23; the ticker symbol is "ENG."

Synthetic Diamond Grit

De Beers Consolidated Mines, Ltd., announced that it has arranged for the production in South Africa of syntheic diamond grit on a commercial scale. "At the present time there are ample stocks of natural diamond grit for the world market. In view, however, of the interruption of diamond mining operations in the Congo, the production of synthetic material is being undertaken as a precaution to ensure that there will be no break in supplies to the market."

Drakenfeld & Co. Expansion

After having completed an 18,000 sq. ft. warehouse unit last summer and with a laboratory expansion unit now under construction, the B. F. Drakenfeld & Co., Inc., last month broke ground for a 24,400 sq. ft. addition to their Plant B in Washington, Pa.

GCMI Membership Meeting

The semi-annual membership meeting of the Glass Container Manufacturers Institute will be held at the Boca Raton Hotel, Boca Raton, Fla., from Monday, November 28th through Thursday, December 1st.

General sessions will be held on November 29th and December 1st, and the semi-annual membership meeting is scheduled for November 30th.

E. M. Terner, Metro Glass, Division of National Dairy Products Corporation, is chairman of the program committee. Other members of the committee are: G. F. Collins, Jr., Liberty Glass Company, and R. H. Hetzel, Armstrong Cork Company.

Research on Bastnasite

Under the sponsorship of the Molybdenum Corporation of America research on the rare earth ore bastnasite will be continued and expanded at the Pennsylvania State University. New uses for the rare earths and the preparation of new rare earth products will be investigated. Rare earths have great potential in the glass, ceramic, metallurgical, electronic and nuclear fields.

David R. Mitchell, associate dean, College of Mineral Industries, and H. Beecher Charmbury, professor and head of the department of mineral preparation engineering, will supervise the research program.

Ground breaking ceremonies at B. F. Drakenfeld's Washington, Pa., plant left to right: E. A. Hinkins, vice president, Tygart-Valley Glass Co.; D. Potts, superintendent; Thomas Carnahan; Frederick Schussler; Garey Dickson, architects; Harold Ray, assistant manager, Clayware Colors Division; Ray Provost, secretary, Chamber of Commerce; John Porter; H. M. Brenner, manager, Oxide Colors Division; Fay Eakin, purchasing agent; H. B. Fulton, assistant manager, Oxide Colors Division, Frank Jansma, manager, Clayware Colors Division; V. H. Remington, vice president in charge of the Washington plant; Ray Andrews, manager, Glass Colors Division; R. W. Sperring, assistant secretary, Chamber of Commerce; Bartley Osborne, member Board of Directors, Washington Industrial Develoment Corp.; J. A. Stewaart, assistant manager, Glass Colors Division; O. T. Fraser; Dave Smith, president, Chamber of Commerce; Thomas Tinmouth, superintendent, Iroquise Glass Co., Quebec, Canada (visitor); O. Sedenka, plant manager Robert Hemmis; William Schaal; Boone Morrison; Pierre Andry, Iroquise Glass Co., Quebec, Canada (visitor).

INVENTIONS AND INVENTORS

Glass and Wool Fiber

Colored Glass Fibers. Patent No. 2,932,587. Continuation of application August 31, 1953. This application September 14, 1956. Issued April 12, 1960. One sheet of drawings; none reproduced. Assigned to L. O. F. Glass Fibers Company by Dominick Labino.

It has been found that water soluble Prussian blue, that is, a water soluble ferric ferrocyanide is ideally suited for providing a distinct coloration on glass fibers.

In the preferred practice of the invention, a water soluble Prussian blue is combined with a phenolic resinous binding solution and is applied to the fibers with the bonding agent.

The solution preferably contains:

	Gallons
Phenol formaldehyde (water soluble)	1.25
Prussian blue mix	1
Water	50

The Prussian blue mix contains:

Prussian blue (water soluble)	grams	270
Oxalic acid	do	35
Ammonia (NH_4OH)	quart	1
Water, sufficient to make 1 gallon		

The oxylate present inhibits the precipitation reaction which normally takes place when phenolic solutions are mixed with Prussian blue.

Preferably the phenol formaldehyde and Prussian blue mix are not mixed until shortly before the solution is applied to the glass fibers because even though the oxylate radical inhibits the unwanted precipitation, deterioration of the solution begins after a period of time.

After the resin has been applied to the fiber and heat cured, there is no longer any likelihood of deterioration, and glass fiber mats prepared in accordance with this invention are stable for years.

There were 4 claims and the following references cited in this patent.

United States Patents

302,544, Conant, July 29, 1884; 2,117,371, Slayter, May 17, 1938; 2,315,329, Hood et al., Mar. 30, 1943; 2,319,142, Lebach, May 11, 1943; 2,582,919, Biefeld, Jan. 15, 1952; 2,688,580, Fingerhut, Sept. 7, 1954.

Plastics and Laminates Formed of Glass Fibers and Epoxy Resins. Patent No. 2,931,739. Filed October 8, 1956. Issued April 5, 1960. No sheets of drawings; none reproduced. Assigned to Owens-Corning Fiberglas Corporation by Alfred Marzocchi and Nicholas S. Janetos.

The invention provides a laminate and plastic formed of the system consisting of an epoxy resin as the adhesive phase, glass fibers as a reinforcement and a coating on the glass fiber surfaces to improve the processing characteristics of the glass fibers and to improve the bonding relationship between the epoxy resins. The coating consists of a saturated polyester resin, a polyvinyl alcohol, a glass fiber lubricant-emulsifier, an organosilicon compound, and polyvinylpyrrolidone. These materials are present in the ratio of 2.0 to 5.0 parts by weight saturated polyester resin, 0.1 to 2.0 parts by weight polyvinyl alcohol, 0.1 to 1.0 parts by weight lubricant-emulsifier, 0.2 to 3.0 parts by weight organosilicon compound and 0.3 to 2.5 parts by weight polyvinylpyrrolidone.

There were 14 claims cited in this patent.

Glass Reinforced Metals. Patent No. 2,932,071. Filed October 19, 1956. Issued April 12, 1960. No sheets of drawings. Assigned to Owens-Corning Fiberglas Corporation by Harry B. Whitehurst.

A method for producing a glass-reinforced lead article comprises coating glass fibers with Wood's metal, intimately associating the Wood's metal-coated glass fibers with molten lead, and solidifying the liquid lead.

Specific examples of suggested uses for metal coated glass fibers include the production of reinforced metal articles by either hot or cold pressing, in suitable dies, of masses of the coated fibers or flakes. Best results, using this technique, have been achieved by hot pressing in a hydrogen atmosphere, although some evidence of welding was noted even with cold pressing. Similar results have been noted when masses of fibers or flakes either bare or metal coated, were sandwiched between sheets of metal foil and subjected to pressure, preferably at elevated temperatures.

Electrically conducting masses have also been produced by fabricating the metal coated fibers into a scrim fabric, and then embedding the scrim in a suitable synthetic resinous material such as polyethylene.

There were 9 claims and the following references cited in this patent.

United States Patents

604,569, Ringstrom, May 24, 1898; 1,118,812, Sand et al., Nov. 24, 1914; 1,778,893, Forrester, Oct. 21, 1930; 2,424,353, Essig, July 22, 1947; 2,699,415, Nachtman, Jan. 11, 1955; 2,772,987, Whitehurst et al., Dec. 4, 1956.

Foreign Patent

171,932, Australia, July 25, 1952.

(*Continued on page* 659)

NEW APPLICATIONS IN GLASS...

Fig. A

Glass Curtain Wall in Building

Factory-fabricated glass tile panels, four feet wide, two feet high and two inches thick, have been used in constructing the nation's first mechanized post office at Providence, R. I.

Six poured-in-place parabolic domes with a clear span of 150 feet make up the building's roof.

The areas beneath the domes' arches (Fig. A.) are comprised of insulating (Thinlite) glass curtain wall which will provide glare-free daylight to the interior of the building. (Fig. B.) The panels are bolted to previously-erected vertical struts installed in the parabolic arch openings.

Incorporated in the glass tiles are special prisms that eliminate glare on direct-sun exposures and reduce heating and air-conditioning costs substantially. The original cost of air-conditioning equipment was cut about $45,000 and the annual fuel consumption during winter months will be reduced by an estimated 17,000 gallons.

Approximately 31,000 square feet of the insulating glass tile panels have been used on the building.

Insulating Glass: Kimble Glass Co., subsidiary of Owens-Illinois, Toledo, O.

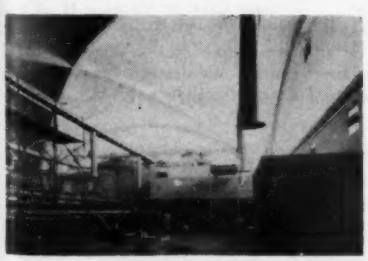


Fig. B

Glass vs. Heat and Humidity

Glass blocks have replaced 85 per cent of the conventional windows and sashes at two of Canton Cotton Mills' plants in Canton, Ga. to help obtain a proper balance of humidity and heat. An improper balance of humidity and heat causes static electricity in the dry cotton, card fly fills the air, and production is halted until correct levels are restored.

Glass Blocks: Pittsburgh Corning Corp., One Gateway Center, Pittsburgh 22, Pa.

Glass Pipe

A lightweight, air-cooled, smoking pipe is equipped with a glass bowl and claimed not to bite, sting, or irritate the smoker. The heat resistant bowl is completely inert and non-absorbent and has no secondary heat. Bowl cover cooling fins and top ring are of nylon compound and stem is nylon.

Kimax glass bowl: Kimble Glass Co., Toledo 1, O.
Pipe: Pyro-Bol Corp., 653 S. Wells St., Chicago 7, Ill.

Special Glasses

Small pillows and balls of glass are available for use in filler, packing, filtering and tumbling applications. Specific applications include: packing in aircraft wings for structural support and buoyance; filler material in the casting of large plastic pieces; fillers in refractionating columns and filter beds for chemical processing; and as anti-lint filters in automatic washing machines.

GLASSES IN POWDER FORM are available for use as a phosphor-containing medium in electroluminescent devices. They are formulated to provide chemical compatibility with electroluminescent phosphors and to match thermal expansion values of glass and ceramic substrates.

DISCS OF POROUS GLASS have been developed as tiny moisture getters for semiconductors. They are placed within semiconductor enclosures because the devices tend to become less efficient in the presence of moisture.

Glasses: Corning Glass Works, Corning, N. Y.

St. Gobain

Gray Windows Cool Building

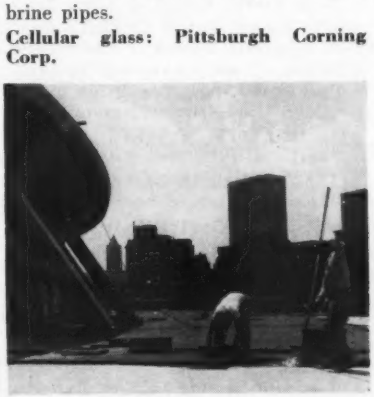
Gray windows capable of reducing the transmission of solar energy were chosen for the new Amity Leather Products building in West Bend, Wisc. The heat absorption properties of glare-reducing glass helps reduce the load on the building's year-round air conditioning.

Glass: American-Saint Gobain Corp., 60 E. 42nd St., New York 17, N.Y.

Cellular Glass

Two-inch thick cellular glass insulation was installed on the podium encircling the outside of Pittsburgh's new Civic Auditorium. A total of 52,000 sq. ft. of the material is being used on the podium and two ramps leading up to it. A second layer of concrete will be installed over the insulation. The podium serves as a roof and as a promenade leading to the auditorium entrances. A high compressive strength insulation was needed to support the weight of the crowds who will walk over it. Cellular glass will also be used beneath the Auditorium's skating rink. It has already been installed in the refrigeration tunnel housing the rink's brine pipes.

Cellular glass: Pittsburgh Corning Corp.



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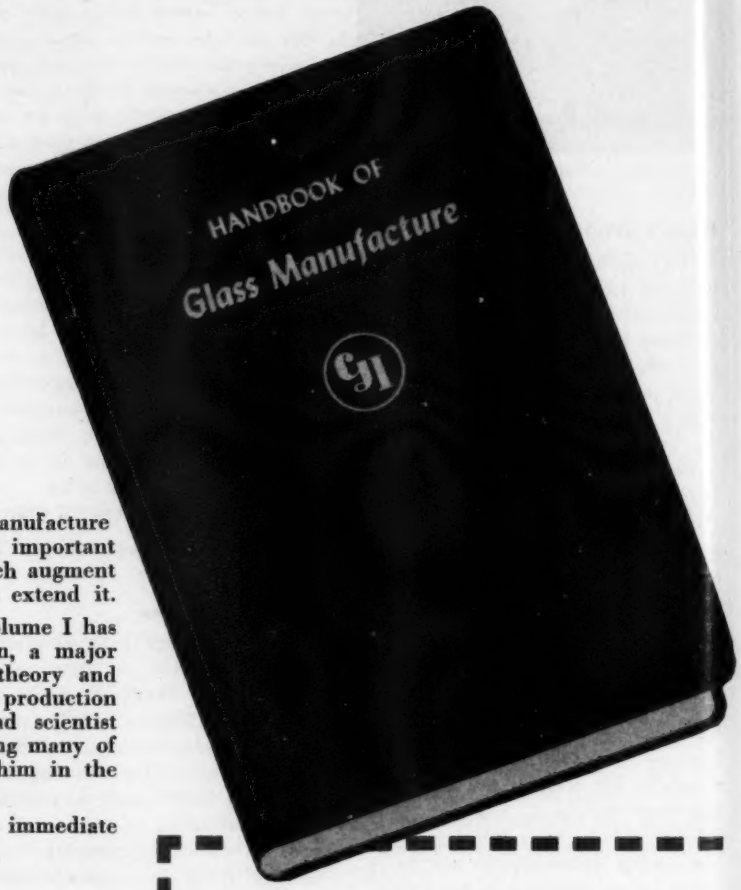

Volume II of the Handbook of Glass Manufacture deals with a number of significant and important topics in the glass industry, some of which augment the subject matter in Volume I, others extend it.

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Torches can be used to finish manufactured glass with a fine polish through surface melting; pierce holes in glass; coat glass with ceramic or metal—from silver to tungsten; solder metal to glass; and spray fixed melting point glasses onto nearly any metallic or ceramic part. The flame is silent and direct impingement on a molten substance causes no appreciable surface disturbance.

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Unit consists of torch, automatic controle console, power supply and all connections. Power levels range from 12 KW to 1000 KW. Descriptive bulletin with performance data, and comparative chart is available.

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Minneapolis-Honeywell, Control Devices Div., 2753 Fourth Ave. South, Minneapolis 9, Minn.

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Device for drop testing of electronic and other fragile equipment in weights up to 150 pounds. Two axial ballbearings guide a 3″ thick high-tensile magnesium casting on a single shaft, reducing friction to a minimum and providing high table rigidity at impact. Available in manual or electrically operated models.

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Medium-capacity glassware washer designed expressly for smaller laboratories. Wide variety of labware can be cleaned, from microscope slides to Erlenmeyer flasks. Operated by steam or electricity and controlled throughout the entire washing program by a selector switch. Rotating drum carries 4 baskets repeatedly in and out of hot wash solution. Water-level regulator prevents power from being turned on until the water is an inch above the heater. Washer contains a 15-gallon tank and is 36″ high, 35″ wide and 22″ deep.

Fisher Scientific Co., 498 Fisher Bldg., Pittsburgh 19, Pa.

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Clark Bros. Co., Olean, N. Y.

CATALOGS RECEIVED

Thermocouple wire. (12 pages) Catalog on thermocouple wire and extension wire, bare and insulated. List prices and ordering directions are included.
MINNEAPOLIS-HONEYWELL REGULATOR CO., Wayne and Windrim Aves., Philadelphia 44, Pa.

Commercial glasses. (16 pages) Revised edition of publication "Properties of Selected Commercial Glasses." Covers thermal stress, heat transmission, electrical properties, corrosion resistance and viscosity data. Includes temperature chart for converting Fahrenheit degrees to Centigrade degrees and vice-versa.
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HARBISON CARBORUNDUM CORP., Falconer, N. Y.

Pressure gauges. (6 pages) Bulletin lists pressure gauges of the bourdon-tube type; thermometers and control instruments. Illustrated.
UNITED STATES GAUGE, Division of American Machine and Metals, Inc., Sellersville, Pa.

Refractories. (4 pages) Detailed descriptions of types and classes of refractories used in chemical and petrochemical industries.
HARBISON-WALKER REFRACTORIES Co., 307 Fifth Ave., Pittsburgh 22, Pa.

Flat and Sight glass. Two product bulletins. One covers flat glass products used to observe flames in furnaces and boilers; study corrosive processes; and shield against infrared rays. Other bulletin gives property data and installation information on sight glasses used in ovens, absorptions columns, reaction kettles, furnaces, stills, and processing tanks.
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Equipment Bulletin. (12 pages) Illustrated. Discusses applications and performance characteristics of pneumatic conveyors, rotary compressors and vacuum pumps, suspension-type preheaters, positive displacement blowers and gas pumps, and induced draft fans.
FULLER CO., Catasauqua, Pa.

Firebrick. (4 pages) For high resistances to slagging and abrasion. Includes technical data on pyrometric cone equivalents, hot load deformation, permanent linear change, panel spalling, and porosity. Chemical analysis of each type of brick is also included.
J. H. FRANCE REFRACTORIES CO., 9360 France Rd., Snow Shoe, Pa.

CURRENT STATISTICAL POSITION OF GLASS

Employment in the glass industry during July, 1960, was as follows: *Flat Glass:* a preliminary figure of 25,800 for July, 1960, indicates a decrease of 1.5 per cent under the adjusted figure of 26,200 reported for June, 1960. *Glass and Glassware, Pressed and Blown:* a decrease of 3.7 per cent is shown by the preliminary figure of 89,700 reported for July, 1960, when compared with the adjusted figure of 93,200 reported for June, 1960. *Glass Products Made of Purchased Glass:* the preliminary figure of 13,300 given for July, 1960, is 2.2 per cent under the adjusted figure of 13,600 reported for June, 1960.

GLASS CONTAINER SHIPMENTS

(All Figures in Gross)

Narrow Neck Containers

	August, 1960
Food	2,225,000
Medicinal and Health Supplies	1,554,000
Chemical, Household and Industrial	1,063,000
Toiletries and Cosmetics	1,103,000
Beverage, Returnable	678,000
Beverage, Non-returnable	170,000
Beer, Returnable	181,000
Beer, Non-returnable	2,118,000
Liquor	846,000
Wine	421,000
Sub-total (Narrow)	10,359,000

Wide Mouth Containers

Food	*4,638,000
Medicinal and Health Supplies	446,000
Chemical, Household and Industrial	179,000
Toiletries and Cosmetics	303,000
Dairy Products	201,000
Sub-total (Wide)	*5,767,000
Total Domestic	16,126,000
Export Shipments	*186,000
TOTAL SHIPMENTS	*16,312,000

* This figure includes Fruit Jars, Jelly Glasses, and Packers' Tumblers.

GLASS CONTAINER PRODUCTION AND INVENTORY

(All Figures in Gross)

		Production August 1960	Stocks August 1960
Food, Medicinal and Health Supplies; Chemical, Household and Industrial; Toiletries and Cosmetics	Narrow Neck	5,547,000	7,771,000
	Wide Mouth	*5,541,000	*7,226,000
Beverage, Returnable		760,000	2,298,000
Beverage, Non-returnable		167,000	330,000
Beer, Returnable		200,000	473,000
Beer, Non-returnable		1,925,000	734,000
Liquor		953,000	1,576,000
Wine		400,000	751,000
Dairy Products		178,000	341,000
TOTAL		*15,671,000	*21,500,000

* This figure includes Fruit Jars, Jelly Glasses, and Packers' Tumblers.

Payrolls in the glass industry during July, 1960, were as follows: *Flat Glass:* a decrease of 1.8 per cent is shown in the preliminary $13,961,627.95 given for July, 1960, when compared with June's $14,223,497.13. *Glass and Glassware, Pressed and Blown:* a decrease of 4.2 per cent is shown in the preliminary $35,913,117.24 given for July, 1960, when compared with the previous month's adjusted $37,500,173.82. *Glass Products Made of Purchased Glass:* a preliminary figure of $4,311,794.30 was reported for July, 1960. This is a decrease of 0.7 per cent when compared with the adjusted figure of $4,343,641.85 for June, 1960.

Glass Container Production based on figures released by the Bureau of the Census, Industry Division, was 15,671,000 gross during August, 1960. This is an increase of 10.4 per cent over the previous month's production figure, 14,185,000 gross. During August, 1959, glass container production was 15,019,000 gross, or 4.1 per cent under the August, 1960 figure. At the end of the first eight months of 1960, glass container manufacturers have produced a preliminary total of 110,463,000 gross. This is 5.1 per cent more than the 105,057,000 gross produced during the same period in 1959.

Glass Container Shipments during August, 1960, came to 16,312,000 gross, an increase of 27.8 per cent over July, 1960, which totaled 12,762,000 gross. Shipments during August, 1959, amounted to 20,116,000 gross, or 23.3 per cent over August, 1960. At the end of the first eight months of 1960, shipments have reached a preliminary total of 106,075,000 gross, which is 0.6 per cent less than the 107,706,000 gross shipped during the same period of the previous year.

Stocks on hand at the end of August, 1960, came to 21,500,000 gross. This is 2.5 per cent under the 22,061,000 gross on hand at the end of July, 1960, and 56.3 per cent more than the 13,748,000 gross on hand at the end of August, 1959.

NEW BOOKS

SOME MATHEMATICAL METHODS OF PHYSICS, by Gerald Goertzel, Technical Director, and Nunzio Trailli, Head, Theoretical Physics Section, Nuclear Development Corporation of America. McGraw-Hill Book Co., Inc., 330 W. 42nd St., New York 36, N.Y. 300 pages. $8.50

Publication gives practical mathematical methods for solving problems in such fields as electromagnetic theory, quantum mechanics, reactor physics and acoustics. The techniques illustrate the basic concepts involved in the study of linear systems with particular emphasis on eigenvalues, eigenfunctions, and Green's functions. Among the subjects covered are Fourier analysis, perturbation, variation, and numerical approximation methods; matrices; operators; vector spaces and orthogonal functions.

CURRENT REVIEW OF THE SOVIET TECHNICAL PRESS. Office of Technical Services, Business and Defense Service Administration, U.S. Department of Commerce. Sold on subscription, $7.00 for six months.

A weekly review of scientific and technical articles appearing in the Soviet press and of significant new Russian books. The time lapse between appearance of the original article and its review is said to be no more than a month. Articles are not translated in their entirety but each review cites the original Russian publication including page unmbers.

The reviews cover articles in a broad field of science and technology, including ceramics, electronics, use of ultrasonics, mechanical engineering, metals, radioactivity, and radiation in industry and activities in space.

ZIRCONIA: ITS CRYSTALLOGRAPHICPOLYMORPHY AND HIGH TEMPERATURE POTENTIALS, by B. C. Weber and M. A. Schwartz, Aeronautical Research Laboratory, Wright Air Development Center, U.S. Air Force. Order PB 151665 from Office of Technical Services, U.S. Department of Commerce, Washington 25, D.C. 26 pages, 75 cents

This report clarifies some of the controversial data presented in literature concerning this refractory metal, reviews the chemistry of zirconia, and presents some of its future potentialities. Report states that the usefulness of zirconia is now dependent on controlling its crystallographic transformation. Also describes the advancements in "metal-modified oxides."

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INFRARED RADIATION, by Henry L. Hackforth, Nortronics, Division of Northrop Corp. McGraw-Hill Book Co., Inc., 330 W. 42nd St., New York 36, N.Y. 288 pages. $10.00

Describes infrared radiation technology and its role in optics, electronics, medicine, biology, space science, astronomy, meteorology, geophysics, and many other fields. A generalized infrared system is developed to aid in understanding. The changes undergone by a signal from its source, through the system, to the final display are shown. Information on components, the laws of physics by which they operate, sources of radiation, methods of transmission, and the analysis and design of systems is included. Material is also presented on infrared artificial sources; research in wavelength transmission; new optical materials and system, recent types of detectors; and new types of infrared instruments.

TEN LARGE SCALE PHASE EQUILIBRIUM DIAGRAMS, by E.F. Osborn and Arnulf Muan, College of Mineral Industries, The Pennsylvania State University. Published by The American Ceramic Society, 4055 North High St., Columbus 14, O. $15.00

Printed on 19½ x 23¼-inch sheets of map paper, the diagrams are on a 500 mm. equilateral triangle, with a scale accurate enough to permit interpolation to 0.1 per cent. The base triangle, printed in light blue, has 5 mm. division, each representing 1 per cent in composition. Temperature contours are in green, compositions in red, and boundary curves and primary phases in black. Each diagram lists principal literature references to provide sources for further information. The series is listed below:

1. CaO-Al_2O_3-SiO_2
2. CaO-MgO-SiO_2
3. MgO-Al_2O_3-SiO_2
4. NaO-Al_2O_3-SiO_2
5. K_2O-Al_2O_3-SiO_2
6. FeO-Fe_2O_3-SiO_2
7. CaO-FeO-SiO_2
8. MgO-FeO-SiO_2
9. FeO-Al_2O_3-SiO_2
10. CaO-Fe_2O_3-SiO_2

METAL FIBER REINFORCED CERAMICS, by R. S. Truesdale, J. H. Swica, and J. R. Tinklepaugh, Alfred University for Wright Air Development Center, U.S. Air Force. Order PB 151610 from Office of Technical Services, U.S. Department of Commerce, Washington 25, D.C. 45 pages. $1.25

Techniques are described for the sintering and hot pressing of alumnia and aluminas containing 5, 10, and 20 percent (by weight) molybdenum fibers. Physical and mechanical properties of alumina containing the ⅛-inch long by 0.002-inch diameter fibers were determined and compared to those of the alumnia. The alumina was superior in strength and impact-resistance, however, aluminas containing 10 and 20 weight-per cent additions of fiber seemed superior in thermal shock resistance.

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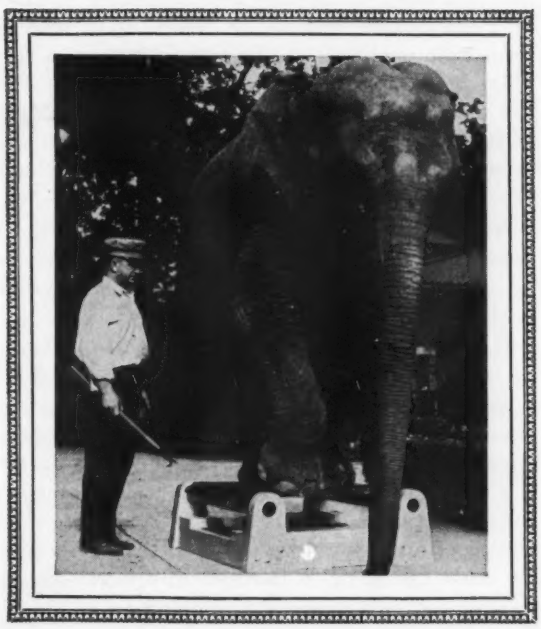

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CONDITIONS OF GLASS FORMATION

(*Continued from page* 627)

systems, a complexity which corresponds to that of an organic high polymer whose synthesis leads to a variety of molecules which differ in molecular weight and in the degree of branching.

L. T. Trostel[46] melted quartz and kept the melt at 1760°C for ten minutes. Under these conditions, which means 50 degrees above the melting point of cristobalite and more than a hundred degrees above that of quartz, the liquid still "remembered" the long range order of the quartz crystals. Devitrification of this melt produced quartz rather than cristobalite. We refer to this type of solid-liquid transition as the melting of a "perfect" crystal because it is controlled by the kinetic energy and the average binding forces in the lattice.

Most substances do not melt this way: most crystals develop defects on heating and the concentration of defects increases very rapidly at a certain temperature so that the lattice breaks down because of the disproportionation of the average binding forces into stronger and weaker forces. All solids develop defects on heating. A lattice defect—in our discussions we used the anion vacancy as an example—raises the potential energy as well as the entropy of the system. The stability of a system is measured by its free energy F and the free energy is related to the potential energy E and the entropy S according to

$$F = E - TS.$$

The formation of a defect raises the values of E and of S, and thus affects the value of F in an antagonistic way. The entropy S appears in this equation multiplied by T the absolute temperatiure. Hence, under otherwise comparable conditions, the formation of a certain defect which raises the potential energy by a certain value and which produces a certain increase of the configurational entropy of the system will make a much more important contribution to its free energy at high rather than at low temperatures.

Because of this entropy contribution the kinetics of melting of the high melting carbides, borides, nitrides, etc. will be "defect controlled." These high melting substances do not melt as "perfect" crystals.

The two antagonistic effects which a defect has on the stability of a system leads to the establishment of a temperature dependent equilibrium. At a given temperature a crystal is stable if it contains the equilibrium concentration of defects. Crystals which contain a greater or a smaller number of defects have a higher free energy and are unstable or metastable. The kinetics of the melting of a substance depends upon the way in which the defect concentration increases with increasing temperature and on the absolute melting point of the substance.

Substances which melt because of a rapid increase of the defect concentration are not likely to form glasses. These crystals cannot be overheated. Their melts, even when supercooled, remain fluid and do not form a glass.

It might be important to briefly state that we are speaking here of structural defects which are the centers of an asymmetry with respect to the binding forces. Recently scientists paid much attention to the electronic defects such as the color centers produced by irradiation. Exposing a glass to sunlight or a quartz crystal to X-rays can produce color centers which are the result of an electron transfer process. These defects have little or no influence upon the melting and nucleation because they heal at elevated temperatures.

According to our views the tetrahedral coordination of the cations is not of primary importance for its ability to form stable glasses. We attribute the high stability of vitreous potassium tetrasilicate not to the tetrahedral coordination but to its very low melting point as compared with other binary silicates. We explain the high stability of a lead orthosilicate glass on the basis of its very low melting point as compared with other orthosilicates. The structure of the potassium tetrasilicate very likely corresponds to the picture which can be derived on the basis of the Zachariasen-Warren concept but the atomic structure of the lead orthosilicate glass does not. We see no justification in singling out tetrahedral coordination as a prerequisite of glass formation.

High melting temperatures impair glass formation for two reasons. Firstly, because high thermal energies are necessary for overcoming the energy barrier of nucleation and, secondly, because at high temperatures the value of TS is high so that defects can contribute more to the stability of a system at high than at low temperatures. Defects are centers of asymmetry which lower the energy barrier of nucleation because they cause disproportionation in the binding forces of the system.

The formation of defects is not limited to metals and ionic crystals. All lattices, even that of phenolphthalein, develop defects if the temperature is raised. A common feature of all glasses is the increase of the number of defects above T_g. In the case of an organic compound, e.g., vitreous phenolphthalein, we cannot speak of vacant anion sites but we can describe the structural change beginning at T_g as a decrease of the average coordination number of the molecules.

Defects are effective in nucleation not necessarily because they represent holes in the lattice which enhance the diffusion but because they are centers of an asymmetry which affect the binding forces over large distances. Thus Na_2O in small concentrations introduces an asymmetry into SiO_2 because it provides O^{2-} ions of high polarizability. H_2O and HF introduce an asymmetry because some singly charged OH^- or F^- ions take the place of some doubly charged O^{2-} ion. Al_2O_3 introduces an asymmetry and so does WO_3 because these oxides introduce cations which have charges which differ from that of the Si^{4+} ion. All of these additions increase the rate of nucleation of vitreous silica.

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INVENTIONS

(*Continued from page* 650)

Glass Compositions

Glass Composition. Patent No. 2,882,173. Filed June 20, 1955. Issued April 14, 1959. No sheets of drawings; none reproduced. Assigned to Owens-Corning Fiberglas Corporation by William W. Welsch.

This invention relates to a glass composition which is especially adapted for the manufacture of glass articles such as fibrous glass and the like.

It has been discovered that a glass comprising silica and the usual glass-forming ingredients plus an addition of boric oxide and a compound containing fluorine provides the physical properties which are desired for producing fibrous glass by a rotary process. The glass compositions of this invention may be utilized in other fiber-

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forming processes in which it is desirable to have a wide range of fiber-forming or attenuating temperatures.

A suitable glass composition comprises the following ingredients expressed in weight per cent: SiO_2, 50 to 65; Al_2O_3, 0 to 8; CaO, 0 to 10; MgO, 0 to 10; Alkali (Na_2O and K_2O), 10 to 20; B_2O_3, 0 to 10; CaF_2, 5 to 15; TiO_2, 0 to 8; ZrO_2, 0 to 8.

There were 8 claims and the following references cited.

United States Patents

2,426,472, Stanworth, Aug. 26, 1947; 2,429,432, Stanworth, Oct. 21, 1947; 2,687,968, Beck, Aug. 31, 1954.

Foreign Patents

897,060, France, Mar. 12, 1945; 905,421, Germany, 1954.

Other References

Locke: THE GLASS INDUSTRY, Vol. 7, No. 6 (1926), page 136.

Miscellaneous Processes

Pressing apparatus for glass laminates. Patent No. 2,825,671. Filed December 22, 1954. Issued March 4, 1958. No sheets of drawings; none reproduced. Assigned to Pittsburgh Plate Glass Company by Victor K. Langhart and Charles O. Huffman.

This invention relates to apparatus for pressing of bent glass laminated assemblies comprising two matched sheets of curved glass and a thermoplastic interlayer, and especially for the preliminary pressing of the laminated assemblies prior to placing the assemblies in an autoclave for completion of the laminating process.

The object is to provide a preliminary pressing apparatus with a pair of yieldable rolls that will be mechanically moved at their ends in horizontal and vertical directions in a predetermined manner so that their positions will anticipate the twist of the glass along its longitudinal axis.

The apparatus comprises a pair of driven nipper rolls mounted in frictional engagement in a rotatable roll housing. The roll housing is mounted to a pair of spaced plates which are movably mounted in relationship to a fixed housing. The spaced plates are moved in vertical and horizontal directions relative to the fixed housing in a controlled and predetermined manner. This is accomplished by means of hydraulic cylinders actuated through cams rotating with the nipper rolls and upon actuation of a trip mechanism by an approaching bent glass laminated assembly. Control of plate positioning as called for by one of the cams is provided by feed-back mechanism actuated by vertical or horizontal movement of a spaced plate.

There were 16 claims and the following references cited in this patent.

United States Patents

2,525,980, Walters, Oct. 17, 1950; 2,601,794, Wood, July 1, 1952.

Siloxane rubber glass cord tire. Patent No. 2,827,090. Filed July 8, 1955. Issued March 18, 1958. No sheets of drawings; none reproduced. Assigned to Dow Corning Corp., by Delmar C. Youngs.

The increased use of heavy load carrying vehicles and of high speed vehicles is placing an ever increasing burden on tires made from organic rubber reinforced with organic cords. For example, it has been found that tires used on heavy trucks tend to delaminate due to the heat build-up inside the tread and between the plies. Another cause of failure in present tires is due to the fusion or deterioration of the cords when subjected to higher temperatures.

This invention relates to a tire comprising silicone rubber reinforced with glass cords, the individual filaments of which are coated with a film of a cured organopolysiloxane having on the average from 1.9 to 2 monovalent hydrocarbon radicals per silicon atom.

The tire of this invention is prepared by coating individual glass filaments with the aforesaid siloxane, spinning the filaments into a cord, embedding the cord in sheets of solicone rubber and thereafter laminating the sheets so formed and molding them into a tire.

The resulting tire is suitable for use on wheeled vehicles particularly when operating at high temperatures, i.e. above 150°F and extremely low temperatures such as −75°C.

There were 2 claims and the following references cited in this patent.

United States Patents

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STATEMENT OF THE OWNERSHIP, MANAGEMENT, AND CIRCULATION REQUIRED BY THE ACT OF CONGRESS OF AUGUST 24, 1912, AS AMENDED BY THE ACTS OF MARCH 3, 1933, AND JULY 2, 1946 (Title 39, United States Code, Section 233)

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Commission Expires March 30, 1962.

2,492,129, Spring, Dec. 20, 1949; 2,504,388, Braley, Apr. 18, 1950; 2,749,960, Schwartz, June 12, 1956.

Apparatus and method for edge grinding of plates. Patent No. 2,826,872. Filed November 1, 1954. Issued March 18, 1958. No sheets of drawings; none reproduced. Assigned to Pittsburgh Plate Glass Company by Bruce D. Robbins.

This invention relates to improvements in the method of an apparatus for grinding and finishing the peripheral edge of plates, or panes of shatterproof glass.

In grinding the peripheral edge of glass plates, such as window plates for automobiles, the plates are chucked up and rotated about a center of rotation. A main grinding wheel grinds the peripheral edge in accordance with a given pattern and a finishing wheel effects a finishing polish thereafter.

The primary object of the invention is to control the pressure applied to the finishing wheel so that a greater pressure is applied in certain selected edge portions to insure a higher degree of polish where the plate of glass will be exposed.

This objective is accomplished by providing a cylinder and piston power unit, a spring coupling between the power unit and the finishing wheel, a controlled fluid pressure supply for applying pressure to the power unit and a cam means for controlling the fluid pressure supply to the power unit. The cam means is rotated in synchronism with the rotation of the plate whose edge is being dressed.

There were 8 claims and the following references cited in this patent.

United States Patents

1,575,694, Kuhn et al., Mar. 9, 1926; 2,080,280, Klages, May 11, 1937; 2,084,138, Gottschalk, June 15, 1937.

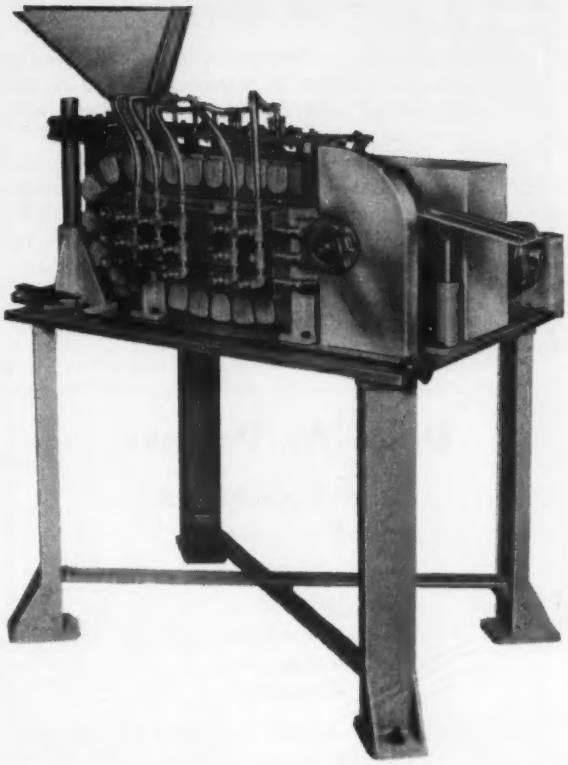
Foreign Patents

966,400, France, Mar. 1, 1950.

Apparatus for optical inspection of glass sheets. Patent No. 2,889,737. Filed November 23, 1954. Issued June 9, 1959. No sheets of drawings; none reproduced. Assigned to Libbey-Owens-Ford Glass Company by Charles E. Griss and Norman V. Huber.

A glass sheet to be inspected is positioned in front of a concave mirror and is illuminated by a brilliant concentrated light source located near the center of curvature of the mirror. An image of the light source as focused by the mirror is received upon a small opaque screen of just sufficient area to intercept all of the undeviated rays of light. If there are areas of the glass being inspected that exhibit wedge, these areas (acting as thin prisms refracting the rays of light) produce deviations of the rays of light both when they pass through the glass going to the mirror as well as when they return from the mirror. The deviations so produced are additive and are sufficient so that the deviated rays of light miss the opaque screen and may be collected and directed toward a photoelectric cell for signalling the presence of wedge in the glass sheet being inspected.

When the deviated rays of light strike the photo-electric cell, they are converted into electrical energy and through various control means actuate a marking device adjacent the glass sheet being inspected and which serves to indi-



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